A MANUAL OF
RADIOACTIVITY

BY

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AND

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COMPLETELY REVISED AND ENLARGED

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PREFACE TO THE SECOND ENGLISH EDITION

THIS second edition of our *Manual of Radioactivity* is the fourth version of the book. It appeared originally in German (1923), then in English translation (1926), then in a revised German edition (1931), and now it appears again in English. On each of these occasions we have endeavoured to bring the book thoroughly up to date, and to retain its essential characteristics.

Since the publication of the first edition, the need for a general text-book of radioactivity seems to have increased rather than to have diminished. Rutherford's *Radioactive Substances and their Radiations* was published in 1912, and his later work *Radiations from Radioactive Substances*, published jointly with Chadwick and Ellis in 1930, gives a new and detailed treatment of selected chapters on the physical side of the subject only. Meyer and Schweidler's comprehensive volume *Radioaktivität* was last published (2nd edition) in 1927, and we understand that a new edition is not contemplated in the immediate future. Mme P. Curie's extensive treatise *Radioactivité* most nearly approaches in its lay-out the type of book we had in mind when writing the present volume. It appeared in 1935, after her decease, but the treatment of many parts of the book belongs to an earlier date. Hitherto, no other authors appear to have written a text-book in which the attempt is made to treat within reasonable limits the entire field of radioactivity, both in its chemical and in its physical aspects, and in its relation to neighbouring sciences.

The general plan of the book has remained unchanged, but in detail a great deal of it has had to be altered, and several chapters entirely rewritten. We have retained its characteristics as a text-book by dealing fully with the fundamental experiments and theories, and by presenting them in as intelligible a manner as possible. In the consideration of the more recent developments we have often had to make use of a more concise treatment, so as to be able more or less to cope with the abundance of the available material. In this respect the essential purpose of our book will have been served if we have been able to give the reader a general survey of modern developments, and direct him to the study of special works of reference and scientific journals. The references given at the end of each chapter should prove useful in this connexion. We have endeavoured as often as possible to direct attention to authoritative books and to reviews in scientific periodicals; individual papers have only been cited in so far as they justify the inclusion of particular statements in the text, and the absence of references to other papers in no way indicates an expression of opinion on their value, for there are obvious practical limitations to the number of references that can be included in a book of this kind. If we were to attempt to give a complete list of references even for the more recent work, it would be necessary to supplement each chapter by several pages of references, for the amount of published work has grown enormously in recent years, especially in the field of radioactivity. From information that we have received from Professor. St. Meyer, some 1,200 papers on the subject appeared during the year 1936 alone.

Uncertainty in regard to the values of the fundamental constants (electron charge, Loschmidt's number, etc.) gave rise to some difficulty. In view of this uncertainty we have, when permissible, made use of approximate numerical values in the text, and in a table in the Appendix we have listed those which, in the present state of knowledge, are to be regarded as the most probable exact values of the constants.

It is our pleasant duty to record our thanks to Drs. O. Frisch and J. C. Jacobsen (Copenhagen) for their critical perusal of parts of the manuscript, to Professor Stefan Meyer (Vienna) for kindly looking over the proofs and for numerous valuable suggestions for improving the text, and above all to Dr. R. W. Lawson (Sheffield) for his very careful translation and invaluable assistance in various ways during the preparation of this edition of our book.

During the printing of this edition the announcement was made of the lamented death of Lord Rutherford. To him, more than to any one else, the scientific world is indebted for the development of the science of Radioactivity throughout its history. For our own part we should like to take this opportunity of expressing our deep feeling of reverence and gratitude to the late Lord Rutherford for the manifold inspiration and encouragement he gave us during the course of the last twenty-five years.

March 1938.

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FROM THE PREFACE TO THE FIRST GERMAN EDITION

The purpose of the present volume is sufficiently indicated by its title; it is intended to be a text-book of radioactivity.

There already exist several exhaustive works on radiology for the advanced student and the research worker, and detailed monographs on certain branches of the subject have been published from time to time. Nevertheless, it has been our experience, and doubtless many of our colleagues have been faced with the same difficulty, that students—in physics, chemistry, and medicine—invariably ask the question as to which is the most suitable book from which to get an insight into the science of radioactivity at first hand, and without assuming an elementary knowledge of the subject. Owing to the wide range of the subject, this is by no means an easy question to answer.

In view of this fact, we have attempted to select and arrange the subject-matter so as to give prominence to the didactic points of view, somewhat in the same manner as has long been done in every text-book on the related sciences of physics and chemistry. Whereas in the early days of radioactive research, before the relation between the numerous remarkable observations was recognized, it was convenient to make use of the historical development of the subject as an introduction to the new science, we believe that the time is now opportune for completely separating the didactic from the historical development of radioactivity. Our meaning will be clear if we remind the reader that a text-book of chemistry does not begin with a description of the misconceptions of the alchemists, but rather with the much more instructive experiments of a quantitative nature on the composition of water, etc., in spite of the fact that they belong to a much later period and are more difficult to carry out. At the same time we do not consider the historical development to be without interest, and students who are already familiar with the elements of the subject will find in the concluding chapter a résumé of progress in radioactive research from its inception onwards, together with an indication of the part played in that work by the various research laboratories.

We have only introduced the principles of the methods of measurement used in radioactivity, and not the practical manipulative instructions; here again we find justification in the accredited procedure of text-books on physics and chemistry, which leave technical laboratory details to the treatises on practical physics, or analytical and preparative chemistry.

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I

INTRODUCTION

1. Fundamental Experiments

The science of radioactivity derives its name from the chemical element radium, which was discovered in the year 1898, and it is mainly with this substance that the unique phenomena have been observed, the study of which forms the basis of this new science. Radium is obtained by separating in the form of chloride the slight traces of barium contained in uranium minerals, and then subjecting this to fractional crystallization. Under such treatment, the more difficultly soluble fractions become richer in the chloride of radium, which in this manner can finally be obtained in the chemically pure state.† From the chemical viewpoint the salts of radium (chloride, bromide, carbonate, sulphate, etc.) and metallic radium bear a strong resemblance to the corresponding compounds of barium and to metallic barium respectively, but they differ from this and from practically all other chemical elements by virtue of certain special properties. We shall discuss the most important of the experiments which show this peculiarity of radium, but at this stage we shall refrain from the discussion of the experimental difficulties which some of them offer. In these experiments we may use a few centigrams of radium chloride, contained in a small glass tube with sealed ends.

(1) If the glass tube be laid for a few seconds on a photographic plate, then after development the plate is found to be strongly blackened where the tube was situated; at greater distances the effect is less marked.

(2) A screen coated with barium platino-cyanide, such as is used in detecting the presence of X-rays, is also found to become luminous when the tube containing radium is brought near it.

(3) A charged electroscope loses its charge when the tube is situated near it.

(4) Very small quantities of two different gases, helium and radon, gradually accumulate within the tube, and can be detected by the methods of micro-gas analysis.

(5) If part of the glass tube be exceedingly thin, but nevertheless quite airtight, the appearance of the gas helium can also be observed outside of the tube.

(6) The tube has always a slightly higher temperature than its surroundings.

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† Further details of the mode of preparation are contained in Chap. XXII.

In the explanation of these remarkable properties we are especially aided by experiment (4). By means of the most accurate chemical analysis we are able to detect in the tube initially only the two elements radium and chlorine in the form of the solid compound $RaCl_2$. After the lapse of a few days or weeks, however, the tube contains an exceedingly small quantity of a gaseous mixture, the spectroscopic examination of which, in addition to helium, reveals the presence of an element previously unknown, and called radon. Owing to the characteristic nature of its spectrum, and because of its inability to take part in chemical reactions, this element, like helium, must be included among the rare gases. Two new elements have thus been derived from radium chloride, and since the same results can be obtained by using radium bromide, radium carbonate, etc., and metallic radium, we are led to the conclusion that it is the element radium that has been transformed into the other two chemical elements.

A quantitative investigation supplies us with a clue to the manner and nature of this transformation. If we were to seal off 1·3138 gm. $RaCl_2$ (containing 1·000 gm. of the element radium) in an evacuated tube, we should be able to establish by the methods of micro-gas analysis that after four days 0·33 $mm.^3$ of radon have been produced (measured at 760 mm. pressure and at 0° C.), whereas the amount after eight days would be 0·50 $mm.^3$, and after thirty days 0·65 $mm.^3$ Moreover, we should find that no matter how long we wait, even after several years, this last value would not be exceeded. In the case of helium, on the other hand, we should find that if we neglect the growth during the first few days, the accumulation of helium would be proportional to the time that had elapsed since the tube was sealed off, so that after one year we should find approximately 172 $mm.^3$ of helium, after two years twice this amount, and so on.†

This difference in the behaviour of helium and radon must find its explanation in the nature of the two gases. In order to investigate this point we shall suppose them to be collected separately, and observe their behaviour during the course of an extended period of time. The helium is found to remain unaltered, and no matter how long we keep it, the volume remains constant. The behaviour of the radon, however, is quite different. Initially it was completely free from helium, but after the course of a few days we are again able to detect the presence of this gas in the radon, whereas the amount of radon constantly diminishes. After the lapse of four days only one-half of it is left, after eight days only about one-quarter, and after one month only about

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† The above relation does not hold quite accurately, owing to the fact that in the course of several months an appreciable quantity of polonium will have been formed, and the generation of helium from this source must also be taken into account.

one-thousandth of the original quantity of radon is left. This process is accompanied by another surprising phenomenon, which is of particular importance in the interpretation of the process: a deposit of lead is found to form on the walls of the vessel in which the radon has been stored. The quantity of lead formed bears a simple relation to the amount of radon that has disappeared; in place of each atom of the latter an atom of lead makes its appearance, and hence the quantity of lead increases during the first four days to one-half of its maximum amount. After one month the amount of lead remains practically constant.† Furthermore, it can be shown that the tube containing radon gives rise to all the phenomena described under experiments (1), (2), (3), (5), and (6), which we were able to observe with the tube containing radium.

2. The Hypothesis of Atomic Disintegration

Without exception it has been found possible to explain all the above-mentioned observations and experiments by means of a theory which also embraces many other related phenomena not touched on here, and which rendered possible the correct prediction of phenomena not yet discovered. The theory is based on a single hypothesis, in principle quite novel, namely, the assumption of the disintegration of the chemical atoms.

Before the discovery of the radioactive elements it had been possible to attribute the whole of the phenomena in chemistry and physics to the behaviour of the atoms and of their association in the form of molecules. In spite of numerous experiments, science had never succeeded in establishing the transformation of one chemical element into another, and since we regard the atoms as the carriers of the properties of elements, we were forced to the conclusion that chemical atoms remain unaltered under all circumstances. This conception does not stand in contradiction to the fact that, in many processes such as the emission of light, ionization, and especially the formation of chemical compounds, certain properties of the atoms disappear, and others take their place. We need only assume that the essential nature of the atom is maintained, and that only relatively minor changes take place, which can always be made retrogressive.‡

Since the new elements helium and radon are formed from radium, we must assume in this case that atoms of helium and of radon are produced from the atoms of radium. Here we are confronted with a

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† Later on we shall deal with the properties of the lead so produced. It is called radium D.

‡ We shall have occasion in what follows to discuss what we have to understand by the 'essential nature of the atom', in the light of more recent theory.

disintegration of the radium atom, and this process is not reversible. In spite of experiments on these lines, it has never been possible to detect the formation of radium from helium and radon. This property of disintegration is associated not only with radium, but also with radon, as mentioned above, which is transformed in its turn into other elements. The same feature characterizes a large number of other radioactive substances, with which we shall become acquainted later on. Previously established conceptions in chemistry and physics were of no avail in the face of these facts, and it became necessary to develop a new science, the science of Radioactivity.

Like the word radium, the term radioactive was chosen in this new science because the first phenomenon to receive attention was the emission of particularly characteristic rays (*radius* = ray) by such substances; not until later was it noticed that these rays solely represent phenomena accompanying atomic disintegration, and for this reason we now define a substance as being radioactive when the atoms of which it is composed disintegrate spontaneously, and regardless of whether or not the emission of rays can readily be detected in the process.

The phenomena that were mentioned in connexion with our basal experiments (1) to (6) all represent effects produced by these rays; we shall be made familiar with the nature of the rays in later chapters specially devoted to this aspect of the subject. At this stage it will suffice to mention that they are not only useful for the qualitative detection of radioactive substances, but they can also be utilized with especial advantage in the quantitative measurement of such substances. The intensity of each type of ray is simply proportional to the available quantity of the substance emitting it. Instead of the above-mentioned chemical or gas-analytical method of detection of the quantity of a radio-element, which is in practice a very irksome business, we almost always make use of the measurement of its radiation in the determination of the amount present. For this reason it is imperative that we should have an accurate knowledge of the rays, and of the methods used in their measurement.

3. The Rays emitted by Radioactive Substances

In order to render this section more readily intelligible, it will be of advantage to call to mind the different theories that have been formulated as to the nature of rays of light, and other types of rays familiar to physicists. Whereas Newton assumed that rays of light consist of a swarm of very quickly moving material particles, Huyghens's conception was that we are not dealing with the propagation of material constituents, but with that of a state of motion. According to his theory, which, after much tedious discussion and numerous experiments, was found to be more fruitful in the case of light than Newton's theory, we have to regard a ray of light as consisting of the propagation of a state of vibration in the aether. Later, the same two alternatives presented themselves in the case of the cathode rays and X-rays. Here again both modes of explanation found distinguished protagonists, until the emission theory held the field for cathode rays, and the wave theory for X-rays.

For the radioactive substances it is not possible to come to a simple decision one way or the other, for here we meet with both types of rays, some of them of a material nature and others resembling light. The α- and β-rays are corpuscular, whereas the γ-rays, like the closely related X-rays, are simply a form of light of very short wave-length. But it should be mentioned that the developments of theoretical and of experimental physics in recent years have led to the conclusion that a rigid differentiation between the two types of rays is not possible. Even an apparently so typical corpuscular radiation as that of the α-particles, which when collected yield the gas helium, can be regarded in other respects as a wave radiation capable of interference. In a preliminary survey of the subject of the rays from radioactive substances, it is however advisable to retain the differentiation previously given.

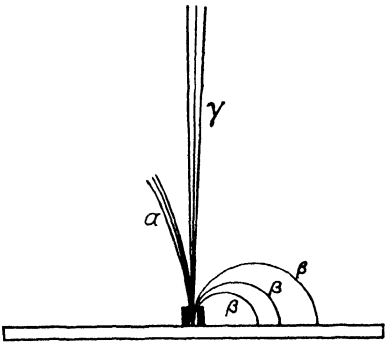

FIG. 1. Action of a Magnetic Field on α-, β-, and γ-Rays.

It has been mentioned that even before the discovery of radium we were familiar with types of rays that were regarded as corpuscular in nature, in contrast to the rays of light. Thus the cathode rays, which result from the application of a high voltage across a highly evacuated tube, consist of a swarm of particles of negative electricity, called electrons; and in the same tube we can detect particles flying in the opposite direction, i.e. towards the cathode, and these are called positive rays. According to the gas contained in the tube (oxygen, carbon dioxide, mercury, etc.), these positive rays consist of molecules and atoms of different chemical elements and compounds, associated with positive electrical charges. β-rays are identical in nature with the cathode rays, whereas the α-rays show a marked similarity to positive rays.

Our knowledge of the nature of the rays emitted by radium was obtained from experiments on their deflexion and absorption. The accompanying diagram (Fig. 1) shows schematically the action of a strong magnet on a pencil of α-, β-, and γ-rays emitted in a rectilinear direction from a narrow opening. In this diagram the magnetic field is supposed to be directed perpendicularly to the plane of the paper, with the north pole of the magnet above that plane. The α-rays are slightly deflected to the left, whilst the β-rays suffer a much greater deflexion to the right. If the magnetic field is sufficiently strong, the β-rays are bent into complete circles, but the γ-rays are wholly uninfluenced by the magnetic field. From the direction of the deflexion we are able to determine whether the charge on the particles is positive or negative, and from the magnitude of the deflexion, combined with similar deflexion experiments in an electric field, we can determine the magnitude of the specific charge (e/m) and the velocity of the α- and β-particles.

Another fundamental experiment consists in testing the absorption of the rays, i.e. the diminution of their intensity after they have passed through matter, whether this be in the gaseous, liquid, or solid state. Whereas the γ-rays are only slightly weakened in intensity by the introduction in their path of an aluminium plate 5 mm. thick, the α-rays are completely, and the β-rays almost completely absorbed by the plate. On the other hand, a thin sheet of paper or of aluminium of thickness 0·1 mm. is insufficient to cut off the β-rays, but it is wholly effective in preventing the passage of α-rays. In this way then we have, together with magnetic and electric deflexion experiments, a simple means of separating the different types of rays from each other.

We must now familiarize ourselves with the methods of measuring the intensity of the rays from radioactive substances. Since these methods are based almost exclusively on the determination of the electrical conductivity imparted to gases by the rays, it will be profitable at this stage to give an account of the mechanism of the conduction of electricity in gases.

4. Conduction of Electricity in Gases

By way of illustration we shall proceed from the more generally familiar conduction of electricity in liquids. As detailed in every text-book of chemistry or physics, pure water is practically a non-conductor of electricity; but when we introduce an electrolyte such as hydrochloric acid into it, the liquid acquires the ability to conduct an electric current. Moreover, it can be shown that at the same time there occurs a wandering of H-atoms towards the cathode, and of Cl-atoms towards the anode. From this it follows that the H-atoms are positively charged, whilst the Cl-atoms possess a negative charge, and that the passage of the current consists in a transport of these charged particles.

Furthermore, we can readily calculate the magnitude of the charge of these particles, which are called ions (*ion* = wanderer). According to Faraday's second law, the same quantity of electricity is always necessary in order to deposit one gram-equivalent of a substance, this quantity amounting to 96,500 coulombs. With this quantity of electricity we can, for instance, separate 1·008 gm. of hydrogen or $\frac{16}{2} = 8$ gm. of oxygen. Now we know that 1·008 gm. of hydrogen contains $6{\cdot}0\times 10^{23}$ atoms, and hence every hydrogen ion bears a charge of

$$\frac{96500}{6{\cdot}0\times 10^{23}} = 1{\cdot}6\times 10^{-19}$$

coulombs. Such small quantities of electricity are advantageously expressed in terms of another system of units; since 1 coulomb $= 3\times 10^{9}$ electrostatic units, it follows that the charge borne by a monovalent ion is $1{\cdot}6\times 10^{-19}\times 3\times 10^{9} = 4{\cdot}8\times 10^{-10}$ e.s.u. A divalent ion requires 96,500 coulombs to deposit half a gram-atom, and since this half gram-atom only contains half the number of atoms contained in a gram-atom, we see that in this case each individual ion carries a double charge $= 2\times 4{\cdot}8\times 10^{-10}$ e.s.u., and that every trivalent ion carries a treble charge, and so on.

The fact that the charge of an ion is either $4{\cdot}8\times 10^{-10}$ e.s.u. or a whole multiple of that value leads us to the atomistic conception of electricity; a smaller quantity of electricity than this elementary charge e has never been observed. As is well known, for these negative 'quanta' of electricity it has been possible to establish their independent existence, free from matter; they invariably possess the same electrical charge, independently of the substance from which they have been derived. They are called electrons, and we shall meet with them again in the β-rays from radioactive substances.

We shall use this well-known conception of the conduction of electricity in liquids as a starting-point in the elucidation of the conduction of electricity in gases, with which the following observations have been made.

Charged bodies that are in contact with a gas gradually lose their charge. Coulomb thought that the gas molecules become charged by contact, and then hand over their charges to the walls of the vessel. But this interpretation stands in contradiction to the observation that the loss of charge is greater the larger the containing vessel. From this we may conclude that the discharge is brought about by the aid of particles which are present in limited number in the vessel, and which cannot be replaced after they have been used up. This conclusion is confirmed by experiments with gases, the conductivity of which is greatly increased by allowing them to bubble through water, by bringing them into contact with flames, electric arcs, glowing metals, or phosphorus, or by subjecting them to the influence of ultra-violet light, cathode rays, positive rays, X-rays, or the rays from radium. The enhanced conductivity vanishes of its own accord in the course of time, but with especial rapidity when the gases are passed through glass wool, through narrow metal tubes, or through an electric field. Thus the conductivity must be attributed to particles that can be filtered from the gases and which are electrically charged. Since the gas as a whole shows no electric charge, equal numbers of positive and negative particles must be present. These particles are also called *ions*, and the production of conductivity is called the *ionization* of the gas.

Thus, just as in salt solutions, the conduction of electricity in gases is brought about by the agency of electrically charged particles. Nevertheless, there are important differences between the two phenomena. In particular, we must note here one such difference, and that is the relation between the applied potential and the current strength. As is well known, this relation is given within wide limits for electrolytically conducting liquids by Ohm's law, $I = V/R$, just as with metallic conductors, i.e. the current is proportional to the potential when the resistance is constant. We explain this relation by saying that with twice the potential the force acting on the ions is twice as large, and hence the velocity they acquire in the resisting medium is also doubled, so that the current or the quantity of electricity transported in unit time will also have double its previous value. This proportionality between current and potential can be graphically represented by means of a straight line passing through the origin. With gases ionized by X-rays or the rays from radium, on the other hand, this simple relation only maintains for the initial part of the curve (region of Ohmic current), and when the potential is sufficiently high a maximum current value is attained, and this cannot under normal circumstances be exceeded.

These conditions can be made clear by graphical means, as shown in Fig. 2.

When we ionize a gas by means of a radioactive preparation of given strength and gradually raise the potential applied to the gas space, the current rises at first quickly, and later slowly, as shown by curve A. The current is constant for potentials above a certain value. With a stronger radioactive preparation the curve has a similar form, but attains a higher constant value (curve B). Thus whereas for small potentials the current strength depends both on the applied potential and on the strength of the radioactive preparation, it is a function only of the strength of the preparation once a certain high value of the potential has been reached or exceeded. We see, then, that under these conditions the current strength can be used as a measure of the strength of the radioactive preparation. This maximum value of the current is called the *saturation current.*

These results are explained in the following manner. Both in the case of electrolytic and of gaseous ions a fraction of the ions disappears by virtue of the recombination of oppositely charged ions to form neutral particles, even in the absence of an electric field. Thus in the aqueous solution of a weak electrolyte in the stationary state a condition of dynamic equilibrium exists between the molecules that are split up into ions in each second of time by the dissociating action of the water, and the ions that reunite during the same interval to form uncharged molecules. Likewise, a state of dynamic equilibrium is also maintained between the gas molecules broken up into ions by the action of the X-rays, and the ions of the gas that combine to form uncharged particles.†

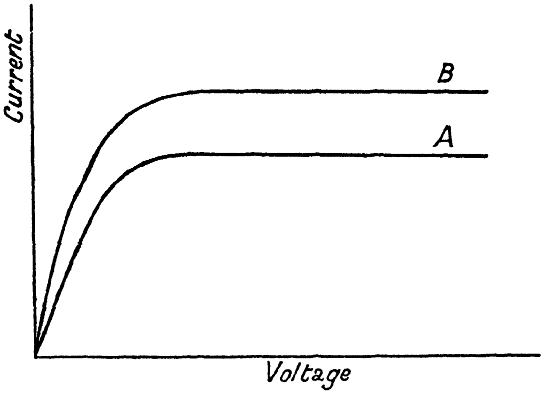

FIG. 2. Attainment of Saturation Current.

When an electric field is applied, the positive ions wander to the cathode and the negative ions to the anode, and if the field is of sufficient strength, the transport can take place so quickly that the ions no longer have an opportunity of losing their charges by recombination, and they all give up their charges to the electrodes. Now with electrolytes this case cannot be realized; first, because the number of ions is very large, and secondly, because they can only be transported slowly towards the electrodes owing to the resistance offered by the water. Thus, even when a strong field is applied, the equilibrium between separation and recombination of the ions is not appreciably disturbed.

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† The velocity of recombination of oppositely charged ions to form uncharged particles is proportional to the product of the concentrations of the positive and negative ions. Hence the number *dn* of the ions that combine per unit of volume in the time *dt* is given by the expression

$$dn/dt = \alpha n_1 n_2.$$

The constant α is called the *coefficient of recombination*. Since the same numbers of positive and negative ions are formed per unit of time, we have $n_1 = n_2$, and the above expression reduces to

$$dn/dt = \alpha n^2.$$

On the other hand, in gases ionized by means of X-rays or the rays from radium the number of ions is much smaller, and their mobility much greater.† In this case, by increasing the potential, it is actually possible to remove to the electrodes all the ions produced during a definite time by the action of the ionizing agent, or in other words, all the ions take part in the transport of the current. As soon as this condition is fulfilled, a further increase of the potential difference is no longer able to bring about an increase in the current, for of course only a definite number of ions is being produced per unit of time. If the ionizing action is increased, the value of the current will also rise, since there are now more ions available. Thus the current is now a measure of the ionizing action, and, within certain limits, it is independent of the potential. Hence, if we wish to measure exactly the ionizing action of a radioactive preparation, it is desirable to make use of the saturation current.‡

The first part of the curves in Fig. 2, the region of Ohmic current, corresponds to the conditions with electrolytes, but the later part of the curves (incomplete saturation and the saturation current) is not attainable in the case of electrolytes.

At first sight it seems a surprising phenomenon that the saturation current becomes greater when the plate electrodes are farther apart; but the explanation of this is also given directly by the theory. If the ionization takes place uniformly throughout the gas, the number of particles produced per unit of time between the electrodes will be larger the greater the distance between the plates, and hence the current may also be larger. In contrast with this we know that for metallic conductors and electrolytes the strength of the current diminishes as we increase the distance between the electrodes.

In view of its practical importance we have dealt in some detail

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† Whereas the ion content of a normal solution of a strong electrolyte amounts to 6×10^{20} per c.c., it rarely exceeds 10^{8} per c.c. in a gas, even with the strongest ionization by X-rays or similar ionizing agency. The mobility of ordinary gas ions in oxygen is about 1 to 2 cm./sec. per volt/cm., and in hydrogen it even reaches the value of 7 to 8, whereas the mobility of most electrolytic ions (Cl, K, etc.) in water only amounts to 7×10^{-4}, and even that of the H-ion does not exceed 3×10^{-3}. Both factors, namely, the smaller number and the greater mobility of the ions, are favourable to the attainment of the saturation current in the case of gases. The conductivity is reduced by the first factor, and increased by the second, but in much lesser degree, so that the conductivity even in hydrogen can only reach a value corresponding to that of an acid solution of strength about 10^{-10} N.

‡ The attainment of the saturation current should be aimed at in all such measurements. It is, of course, also possible to compare the strengths of preparations by using currents below saturation, provided that the conditions of experiment are kept quite constant, and in many measurements we must for practical reasons sacrifice the attainment of complete saturation. Particularly with α-rays, which produce many more ions per centimetre of their path than β-rays (see pp. 43 and 67), it is often not possible to reach saturation.

with the dependence of conductivity on the potential. But there are still other important differences between electrolytic ions and gas ions. In the first place, for the latter ions the close relationship of the charge and the chemical nature of the substance is absent. In electrolysis it is well known that hydrogen always carries one positive charge, and oxygen always carries two negative charges, and so on; but the gas ions in hydrogen and in oxygen carry exactly the same charges. Moreover, whilst we meet with ionized atoms in the solutions of most substances, the gas ions of the same substances generally consist of ionized molecules. In electrolysis all the ions have similar dimensions, even though, in consequence of stronger or weaker hydration, somewhat considerable differences in their magnitude (and corresponding mobility) do occur; but in the case of gas ions the multiplicity is much greater, and at the present time we can visualize the following picture of the process of ionization in gases.

As a result of the collisions of gas molecules with the electrons of cathode or β-rays (or even secondary electrons that have been liberated by X-rays, γ-rays, or ultra-violet light), a neutral particle parts with an electron. As a result of this a positive ion of atomic dimensions is simultaneously produced. At low pressure or at high temperature these ions exist as free ions in the gas, but otherwise uncharged particles attach themselves to the ions by virtue of electrostatic attraction, and so increase the mass of the ions. It has been calculated from diffusion measurements that ordinary gas ions (small ions, or molecular ions) generally consist of a cluster of about thirty molecules. The charge is independent of the number of associated particles; it may remain a single elementary charge, but further charged ions may attach themselves. Apart from electrons, positive atomic ions, and positive or negative molecular ions, 'large ions' (sometimes called 'slow ions') also exist. These deserve mention because they are sometimes present at the commencement of ionization measurements, when they hinder the attainment of the saturation current.

In contrast to the case of electrolytic ions, we must accordingly assume for gas ions an important difference between positive and negative ions; the positive ones are always associated with a mass of atomic order of magnitude, but the negative ions sometimes occur as electrons, particularly when the pressure of the gas is low.

This has an important bearing in connexion with the ionization of gases by collision. We have already seen that for an ionized gas the form of the curve relating the current and the applied potential is represented by Fig. 2, the current remaining constant (saturation current) after a certain value of the potential has been reached. But the value of the current does not remain constant indefinitely as we increase the potential between the electrodes, and at normal pressure a sudden spark ultimately passes between them. If the same experiment be performed with the gas at a reduced pressure of a few centimetres of mercury, we are readily able to determine the exact form of the current-potential curve for the gas, up to the potential at which a luminous discharge takes place. At first it is as in Fig. 2; but when a certain critical potential has been reached, the current again begins to increase, slowly at first, then with increasing rapidity until a glow discharge is initiated. The value of the critical potential depends on the pressure, being less for lower pressures. This renewed increase of the current after the critical potential has been reached is due to the fact that the ions between collisions acquire sufficient energy in the strong electric field to produce further ions by their collision with the neutral molecules of the gas. As mentioned above, when the pressure of the gas is low a considerable fraction of the negative ions exists in the form of electrons, and it is these ions which at first give rise to the growth of the current after the critical potential has been attained; later, however, the positive ions also participate in the process. In this way it is possible to maintain a current of about one thousand times the value of the saturation current. This kind of ionization of a gas, in which the ions initially produced gain sufficient energy by their acceleration in the electric field to produce fresh ions by collision, is called 'ionization by collision'. We shall meet with a very important application of this phenomenon in connexion with the counting of α- and β-particles by means of the point counter and the tube counter (see p. 17).

5. Methods of Measurement for the Rays from Radium

Every well-defined effect of the rays from radium can of course be utilized, in principle, for the purpose of their quantitative comparison. Thus it is very simply shown that radium preparations do not change appreciably in the course of a year, by comparing with each other the blackenings they produce in a given time on a photographic plate (cf. Fundamental Experiment 1), but very exact results cannot be obtained in this manner. Moreover, in quantitative work we should hardly have recourse to the heating effects of radium (Fundamental Experiment 6), since they can be measured accurately only with fairly strong preparations. The best and most general method has been found to be the action of the rays on gases, the insulating power of which for electrical charges is reduced by the rays from radium (Fundamental Experiment 3). As we have seen in the previous section, this phenomenon is based on the ionization of the gases, and can be utilized in a simple manner as the basis of a method of measurement.

Let us suppose that we have a positively charged hemispherical shell of copper, the edge of which rests on a slab of a good insulator, e.g. paraffin wax, and which is surrounded by air. The charge on the hemisphere flows to earth as soon as we touch it with a wire, the other end of which is connected to earth. Normally, air is not capable of conducting electricity, but if we bring a radium preparation near to the charged hemisphere, the air becomes conducting, and electricity now flows through the air in much the same way as if we had brought the hemisphere in contact with an earthed conductor. The air is rendered conducting by the rays from radium, and the degree of conductivity is greater the stronger the radium preparation with which we are working.

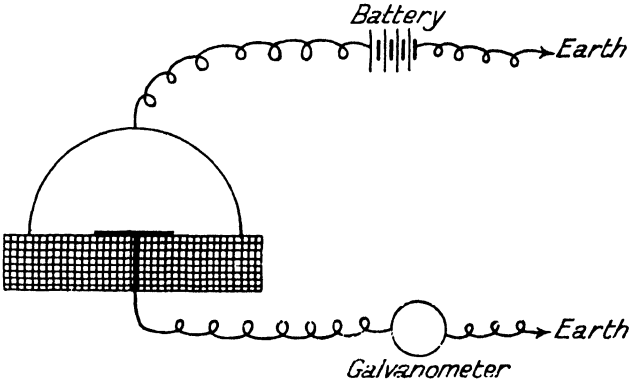

Fig. 3. Apparatus for Measuring Strong Radioactive Preparations.

A slight adaptation of the apparatus just described will serve as an exact measuring instrument. In order to measure the conductivity imparted by the rays from radium to a definite and well-defined quantity of air, we can use that inside the charged hemisphere. As nearly as possible in the middle of the sphere is situated a circular metal plate which rests on the paraffin slab. Connected to this plate is a wire that passes through the slab and is attached to one terminal of a galvanometer, the other terminal of which is earthed, as seen in Fig. 3. The preparation to be measured is laid on the metal plate, the hemisphere is placed over it, and this is connected to a source of potential of from 1,000 to 2,000 volts. If the preparation is not radioactive, the air within the hemisphere will remain an insulator, so that electricity cannot flow between it and the metal plate, and the galvanometer remains undeflected. But if in consequence of the rays emitted by the radium preparation the air between the hemisphere and the plate is ionized, an amount of electricity corresponding to the strength of the ionization will pass from the hemisphere to the plate and through the galvanometer to earth. Thus the current registered by the galvanometer is a direct measure of the ionization produced in the enclosed air, and hence also of the strength of the radium preparation (as shown in section 4), provided the applied potential is sufficiently high to ensure that the maximum or saturation current in the ionized air is attained.

The galvanometric measurements described above can only be carried out with very strong preparations, because the saturation currents obtainable in air are always very weak (see p. 10, footnote), and hence we are obliged to use other more sensitive apparatus, such as is not otherwise used in the measurement of current. For this purpose the electroscope occupies the foremost position. In its simplest form this instrument consists of two strips of metal foil attached at their upper ends; on charging them, the strips diverge. The amount of this divergence is greater the more highly they are charged, and in electroscopes that have been adapted as measuring instruments the divergence of the leaves can be read off on a scale placed behind them. Under normal conditions the leaves remain deflected for a long time, and only gradually collapse ('natural leak'), for the air has very pronounced insulating properties. But if we bring a radium preparation into the neighbourhood of the electroscope, the leaves collapse more or less quickly in accordance with the degree of conductivity imparted to the air by the preparation. If one preparation is twice as strong as another, it imparts to the air twice the conductivity, and hence the leaves collapse at twice the rate, provided that the experimental conditions are maintained exactly the same in the two cases. Thus the time required by the leaves to pass over a given number of divisions of the scale is inversely proportional to the strength of the preparation. Here also the magnitude of the saturation current serves as a measure of the ionizing action. If the volt-calibration of the scale of the electroscope, and the capacity (C) of the entire measuring system, measured in cm., are known, we can calculate the saturation current in the following way: Suppose the leaves collapse from V_1 to V_2 in t seconds, then the drop in potential is (V_1-V_2), and the quantity of electricity that has leaked away is $C(V_1-V_2)$. The electric current, or the quantity of electricity that has passed in one second, is consequently $C(V_1-V_2)/t$. Since 1 volt $= \frac{1}{300}$ electrostatic unit, we must divide the above result by 300 in order to obtain the saturation current in electrostatic units. Our final expression for the current is thus

$$i\,(\text{e.s.u.}) = C\frac{V_1-V_2}{300t}\quad (V_1 \text{ and } V_2 \text{ expressed in volts}).$$

The exceeding simplicity of its use, its rapid action, and its small cost make the electroscope the most favoured instrument in radioactive work.

According to the purpose for which they are primarily required, we differentiate between different types of electroscopes. Fig. 4 shows an

'α-ray electroscope', i.e. an electroscope which, although it permits of the measurement of all three types of rays, has in general too small a sensitiveness for the relatively smaller action of the β- and γ-rays. In this electroscope we have an ionization chamber AB into which the preparation is introduced, and it is separated by an amber or sulphur insulating plug S from the chamber in which the electroscope leaf L is situated. Passing through the plug S and connected with the plate A is a thin metal rod C, which is charged to a definite potential when the instrument is in use, whereas the plate B is directly connected to the earthed outer metal vessel.

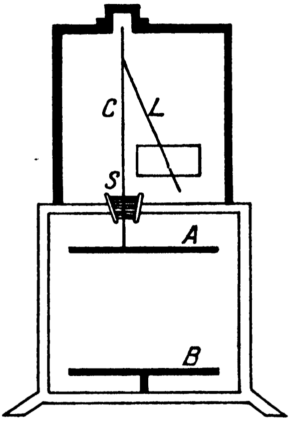

Fig. 4. α-Ray Electroscope.

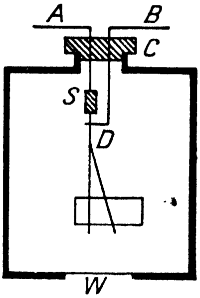

Fig. 5. β-Ray Electroscope.

In 'β-ray electroscopes' (see Fig. 5) the ionization chamber and the measuring chamber are combined, in order to diminish the capacity and hence to increase the sensitiveness. In the base of the instrument there is an opening which is closed only by a thin aluminium foil W; this prevents the α-rays from passing into the instrument, but does not deter the β-rays. The walls of the vessel are of sufficient thickness to prevent the entrance of β-rays. The active preparation is centrally placed beneath the aluminium window. As γ-rays are often present also, they are measured with the β-rays in such a β-ray electroscope, but their effect can generally be disregarded as compared with that of the β-rays. The same electroscope can also be used as a 'γ-ray electroscope' if we close the window W by means of a metal plate of the same thickness as the walls of the electroscope, instead of by aluminium foil. The electroscope is charged through the bent wire B, which rotates in the insulating ebonite stopper C, and can thus be brought into contact either with the earthed case of the instrument or with the leaf-support D below the insulating plug S.

If we require to make measurements with gaseous radioactive substances, we make use of a so-called 'emanation electroscope'. In this instrument the ionization chamber is a closed airtight vessel. Fig. 6 illustrates one of the many possible forms of the instrument, and is sufficiently clear to need no further explanation.

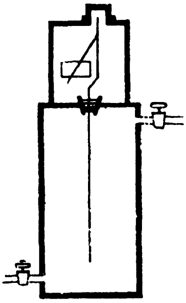
FIG. 6. Emanation Electroscope.

Thin Wollaston wire or fibres of platinized quartz have been found very satisfactory in place of the metallic leaves of electroscopes, especially when we are concerned with precisional measurements. In the 'single-string' (fibre) electrometer the insulated fibre is fixed at both ends and situated between two metal plates connected to the poles of a battery of from 50 to 100 volts, the mid-point of which is earthed. When the fibre receives an electric charge, it is attracted by the oppositely charged plate; the amount of the deflexion gives an indication of the potential acquired by the fibre, and during a measurement the motion of the fibre is observed by means of a microscope with an eye-piece scale. In the 'double-string' electrometer the two adjacent insulated fibres are attached at both ends and repel each other when charged, so that the subsidiary voltage plates are unnecessary. The potential acquired by the system is indicated by the separation of the fibres when charged, and, as before, readings are taken with a microscope with an eye-piece scale.

When working with the rays from a radioactive substance it is in many cases preferable to measure the rate of charging of a previously uncharged electroscope or electrometer, instead of observing the discharge rate with the instrument initially charged. The principle involved in all these methods is the same, however; we are always concerned with the measurement of the saturation current produced in a gas.

In general such instruments are used to establish the presence of radioactive substances, and in these circumstances comparative measurements adequately fulfil our purpose. But if we desire to carry out absolute measurements, we must be able to determine the absolute value of the saturation current, and from what has already been said it will be clear that we must then know the capacity of the instrument and the voltage calibration of the scale of the electroscope. We shall refrain from discussing the methods of measurement in greater detail here, as they are to be found in the special works referred to in the Preface.†

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† For convenience, several different units have been introduced for the measurement of radioactive substances (1 curie, 1 mache, 1 eve, etc.). These different units are dealt with in Chap. XXIII, under the headings of radium and radon.

Finally, special mention should be made of those measuring instruments that enable us to investigate the effects of individual α- and β-particles. The simplest means of recognizing individual α-particles is to make use of a substance which has the property of becoming luminous under the action of the rays from radium (cf. Fundamental Experiment 2, p. 1). Most suitable for this purpose is zinc sulphide which has been prepared in a particular way (see p. 243); this substance is so sensitive towards α-rays that each incident α-particle gives rise to an independent flash of light, which can be seen with the aid of a lens. From the number of these 'scintillations' which appear over a measured area of a screen coated with zinc sulphide, we can calculate the total number of α-particles that have been emitted by the radioactive preparation during the time of observation.

This method was of inestimable value especially in the study of the phenomenon of atomic disruption, since it was found that not only the α-particles, but also the H-particles arising from atomic disruption, call forth scintillations when they are incident on zinc sulphide screens. But it is difficult to exclude the subjective element (visual fatigue) in such observations, and for this reason electrical counting methods are often to be preferred, as they can readily be equipped with an arrangement for the automatic registration of the particles. They possess also the further advantage (sometimes a disadvantage) that they respond not only to α- and H-particles but also to β-particles and even to γ-ray impulses. There exist a variety of forms in which these electrical counting methods have been developed. We shall only discuss the two that are most important at the present time, viz. the point-counter and the tube-counter, both of which were introduced by Geiger.

The general arrangement of the apparatus used with the point-counter can be seen in Fig. 7.

The rays pass through a very thin mica window into a chamber, in the middle of which is situated an insulated metal point or needle. This point is connected to earth through a high resistance, whilst the walls of the chamber are connected to a source of positive high potential. Each time an ionizing particle enters the chamber through the mica window and passes the neighbourhood of the point, a point discharge takes place by virtue of ionization by collision (see p. 11). This produces a sudden deflexion of the fibre of a sensitive string electrometer situated between the point and earth. By suitable choice of the values of the applied potential, the gas pressure and the resistance, the sensitiveness of the instrument can be varied within wide limits, and it can also be made applicable to the registration of individual γ-ray impulses.

When we are concerned with the measurement of a very small flux of radiation, as with the cosmic radiation or the investigation of the artificial radio-elements, the sensitivity of the point-counter is often inadequate. In such cases the tube-counter can be used, since it is very much more sensitive. The tube-counter consists essentially of a metal tube, along the axis of which is stretched a thin wire whose surface is coated with a badly conducting film of uniform thickness. In consequence of the insulating action of this film, the potential between the wire and the tube can be raised beyond the normal spark potential under the prevailing conditions. If at any position inside the tube a small number of ions is produced, the resulting ionization by collision causes an appreciable quantity of electricity to flow to the wire, and this momentary current can be detected with an electrometer, just as in the case of the point-counter described above. Nevertheless, the exceedingly high sensitivity of the tube-counter makes it less suitable than the point-counter for the study of certain problems; for instance, this applies when we wish to detect a weak α-radiation alongside a relatively intense β- and γ-radiation.

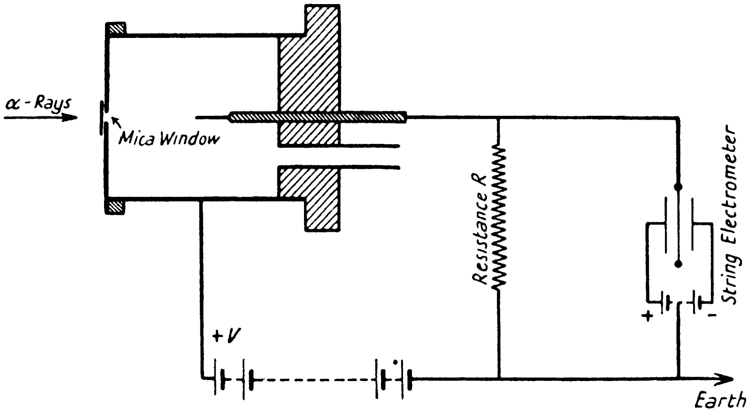

FIG. 7. Geiger's Point-Counter.

A third method which enables us to investigate the action of individual rays is the 'cloud method'. This is based on the fact that supersaturated water vapour condenses preferably on ions. Thus, if by means of a suitable arrangement we produce a supersaturated condition of water vapour in a gas space, in which an ionic track has been produced by a particle of radioactive origin, then in the absence of dust this water vapour will only condense where there are ions, and the track of the particle will consequently become visible as a line of fog. The apparatus used for this purpose was devised by C. T. R. Wilson, and in its earliest and simplest form it is represented diagrammatically in Fig. 8. The movable piston b is suddenly lowered by opening the main valve c, whereby a connexion is established between the vacuum chamber d

and that part of the apparatus situated beneath the piston. This causes an expansion in the space *a*, as a result of which water-vapour supersaturation occurs in the gas content of this space, which is maintained moist by artificial means. If rays enter the expansion chamber in a horizontal direction from a radioactive substance mounted in a side tube of the chamber, these produce ions which are of course at first invisible. However, those ions which are formed during or immediately before or after the expansion serve as condensation nuclei for the water vapour that is in a state of supersaturation at that moment. Tracks consisting of a close array of very tiny droplets are produced in this way, and since the apparatus is closed at the top by a sheet of plate glass, these rows of droplets are visible with suitable illumination as white fog tracks on a dark background. They can also be photographed, and by use of two cameras stereoscopic photographs can be obtained, by means of which the path of the particle in space can be accurately investigated. This method has proved invaluable not only for counting the particles, but also for the study of the fate of the individual particles during their flight. With the aid of this method sudden changes in the direction of the tracks have been detected, and these have led to important conclusions about the collisions occurring between the particles and gas molecules (see Fig. 14, Plate I).

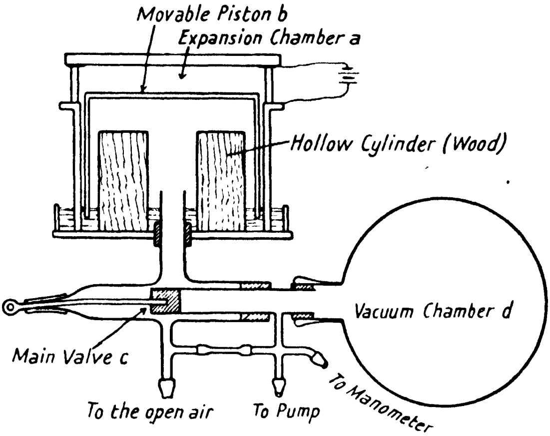

FIG. 8. Wilson's Cloud Chamber.

The recording of individual α-ray tracks in the emulsion of a photographic plate is described on p. 32.

REFERENCE

W. MAKOWER and H. GEIGER, *Practical Measurements in Radioactivity* (Longmans, Green & Co., London, 1912).

II

THE α-RAYS

1. Mass and Charge of the α-Rays

As already mentioned, it follows from the direction of the deflexion experienced by α-rays in a magnetic field that they consist of positively charged particles. They are also deflected in an electric field. If we combine the magnitudes of the electric and magnetic deflexions, we obtain a quantity which is always very characteristic of charged particles, viz. the ratio of the charge to the mass of the particle, or the specific charge, *e*/*m*. We shall consider as the unit of charge the elementary charge of electricity, i.e. that of a mono-valent ion ($e = 4{\cdot}8 \times 10^{-10}$ e.s.u.), and as the unit of mass the mass of a hydrogen atom

$$m = 1{\cdot}67 \times 10^{-24} \text{ gm.}$$

In terms of these units we obtain for *e*/*m* in the case of α-particles the value $\frac{1}{2}$, i.e. such a particle either carries a single elementary charge and has an atomic weight = 2, or it carries two elementary charges and possesses an atomic weight = 4. We are able to decide between these alternatives both by physical and by chemical methods.

A luminescent screen, e.g. a surface coated with zinc sulphide, is caused to emit a flash of light by the incidence of every single α-particle upon it. If we count these scintillations, and determine in addition how much positive electricity is conveyed to an electrometer from the same source of α-rays under identical conditions, we obtain the amount of charge carried by a single particle by dividing the second number by the first. The result is found to be two elementary units of charge. From this it follows that the mass of the particle has the value 4, the atomic weight of helium.

This result is confirmed by the following chemical experiment. If we introduce radon or some other α-rayer into a glass tube with thin walls ($< 1/100$ mm. thick) and seal it off, it is found that the α-particles pass through the tube. But if this tube be now enclosed in a wider evacuated tube with sealed-off ends and thick walls, we are no longer able to detect α-particles outside the latter. The particles must therefore have been retained in the space between the glass tubes or in the walls of the outer one. As a matter of fact, it has been found possible to detect the appearance of helium in the intermediate evacuated space by means of the spectroscope (cf. Fundamental Experiment 5 on p. 1). The amount of helium produced by 1 gm. of radium in one year has the value 172 mm.[3]

We are thus led to the conclusion that α-particles are helium atoms with which are associated two positive elementary charges.

If α-rays are produced during a radioactive transformation, then the process proceeds in such a manner that each disintegrating atom gives rise to one, and only one, α-particle. This important fact was established by means of counting experiments, in which the number of the atoms disintegrating in a given time was known, and the number of α-particles emitted in the same time was determined by the optical method already referred to, or by an electrical method. One of the best electrical methods is based on the fact that a momentary point discharge always occurs from a highly charged needle point when a passing α-particle ionizes the air in the immediate proximity of the point. The method of using this point-counter has already been described on p. 17.

The number of α-particles emitted per second by 1 gm. of radium, free from its products of disintegration, has the value $3{\cdot}7\times 10^{10}$, and when the radium is in equilibrium with three of its disintegration products that emit α-rays, the number is four times as large. From this we can calculate how many helium atoms must be contained in the 172 mm.³ of helium that are produced from 1 gm. of radium together with its disintegration products as far as RaD in the course of a year. This number of helium atoms is clearly

$$3{\cdot}7\times 10^{10}\times 4\times 365\times 86{,}400 = 4{\cdot}66\times 10^{18}.$$

According to the value of Loschmidt's number obtained by quite different methods, there must be present in 1 c.c. of every gas at N.T.P. $2{\cdot}7\times 10^{19}$ molecules, so that theoretically we should expect 172 mm.³ of helium to contain $2{\cdot}7\times 10^{19}\times 0{\cdot}172 = 4{\cdot}65\times 10^{18}$ atoms. This excellent agreement offers convincing proof of the correctness of the atomistic conception of matter, for in the example just given it has been possible literally to count the number of atoms which occupy a definite volume of a gas.

2. Absorption Range and Velocity of the α-Rays

If we bring a preparation which emits α-rays, e.g. a surface coated with polonium, near a zinc-sulphide screen, the latter is found to become luminescent under the action of the rays. On gradually removing the preparation, the luminescence of the screen suddenly ceases at a distance of the radioactive film from the screen which in this case amounts to 3·87 cm. (at 15° C.). The ionizing† and all other actions of the rays also disappear at the same distance (cf. however, the remarks on p. 28).

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† If we introduce the radioactive film into an α-ray electroscope, the two plates of the ionization chamber being separated by considerably more than 3·87 cm., we still find that the preparation produces an effect on the electroscope. Diffusion and transport of the ions of the air in the electric field result in their gaining admittance to the whole of the intervening space before recombination occurs, and hence the electroscope loses its charge.

The existence of a sharply defined limit beyond which no further ionization takes place is a most characteristic property of the α-rays. The determination of this 'range' can be performed with great accuracy by a variety of methods.

In Table 1 are collected together the various radio-elements that emit α-rays, together with the ranges and initial velocities of their α-particles. From the last of these we can calculate the energy of the particles, the resulting value for RaC′ being $7{\cdot}68\times10^6$ electron-volts, which is the energy that an electron would acquire in a vacuum if it fell through a potential difference of $7{\cdot}68\times10^6$ volts. We see that the α-particles from thorium have the shortest, and those from thorium C′ the longest range. The ranges given in the table are the 'practical' or extrapolated ranges, the significance of which is shown in Fig. 11; the ranges of the individual particles fluctuate about a mean range, which is roughly 1 per cent. shorter than the extrapolated range.

TABLE 1

Ranges and Initial Velocities of α-Rays at 15° C. and Atmospheric Pressure

Substance	*Range in cm.*	*Velocity in cm./sec.*
Uranium I [1]	2·63	$1{\cdot}39\times10^9$
Uranium II	3·18	1·48
Ionium	3·19	1·48
Radium	3·39	1·51
Radon (Ra Emanation)	4·12	1·61
Radium A	4·72	1·69
Radium C	4·1	1·61
Radium C′	6·96	$1{\cdot}92_2$
Radium F (Polonium)	3·87	$1{\cdot}59_3$
Thorium	2·59	1·38
Radiothorium	4·02	1·60
Thorium X	4·35	1·64
Thoron (Th Emanation)	5·06	1·73
Thorium A	5·68	1·80
Thorium C	4·78	1·70
Thorium C′	8·62	$2{\cdot}05_2$
Protoactinium	3·67	1·55
Radioactinium	4·68	1·68
Actinium X	4·37	1·65
Actinon (Ac Emanation)	5·79	1·81
Actinium A	6·58	1·89
Actinium C	5·51	1·78
Actinium C′	6·57	1·89

In Fig. 9 (Plate I) are shown the tracks of a very large number of α-particles of normal range from thorium C′; in addition, the track of a

PLATE I

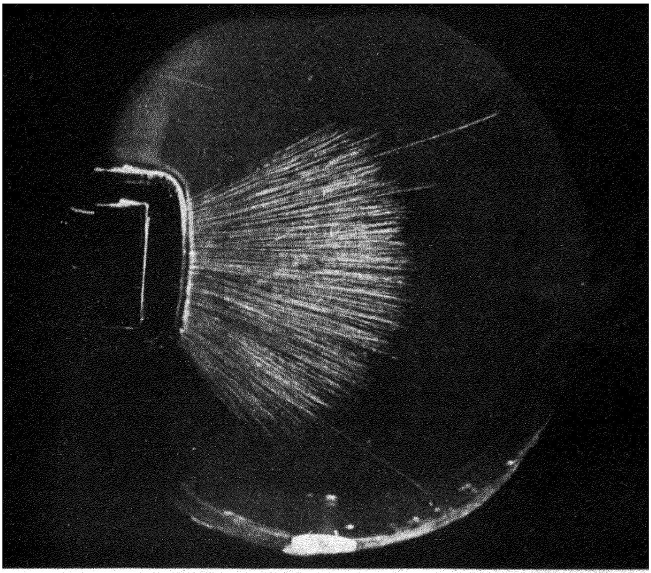

FIG. 9 (p. 22). Tracks of α-Particles from Thorium C′.

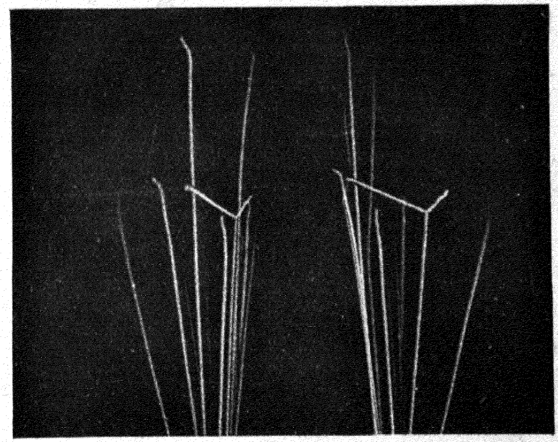

FIG. 14 (p. 32). Deflexion of α-Rays by Oxygen Nuclei.

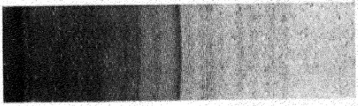

FIG. 17 (p. 40). Magnetic β-Spectrum of the Members of the Actinium Series (RdAc, AcX, AcB, and AcC).

particle of the extreme long range (11·6 cm.) can be seen, as well as that of a particle of 9·8 cm. range (see p. 27).

The range is inversely proportional to the pressure of the gas, and directly proportional to the absolute temperature. Hence α-rays exhibit very large ranges in a high vacuum.

To a first approximation the range is proportional [2] to the cube of the velocity of the α-rays. The velocity, and hence the range of α-particles, can be influenced by the application of strong longitudinal electric fields. Thus the effect of a potential difference of 10,000 volts was to modify the range of the α-particles from polonium in air by 0·226 mm.

Owing to the much greater densities of liquids and solids, the range of α-particles in them is much smaller than in gases. The α-rays from a uranium preparation are absorbed by a sheet of aluminium of thickness 0·018 mm., and the much faster rays from ThC′ by a sheet of thickness 0·065 mm. Accurate range determinations in liquids and in solids are much more difficult to carry out than in gases, but even with them reliable values have been obtained by special methods. For example, Table 2 contains the values found for the ranges of the α-particles from RaC′ in several solid elements. We see that the range diminishes as the density of the material increases. In water, the range of the rays from RaC′ has the value 60·0 μ, and 50·0 μ in the sensitive film of a photographic plate.

TABLE 2

Range of the α-Particles from RaC′ in different Metals [3]

Metal	*Range* (μ)	*Metal*	*Range* (μ)	*Metal*	*Range* (μ)
Li	141	Si	47·2	Ag	19·8
Be	42·8	K	150	Sn	30·1
C	35·1	Fe	19·4	Pt	13·1
Mg	59·5	Cu	18·2	Pb	25·6
		Mica	36·0		

For the purpose of practical measurements a medium can also be conveniently characterized by the statement of its ‘air equivalent’, instead of by the range. This term refers to the thickness of a layer of air which would reduce the range of an α-particle by just the same amount as the medium in question. Mica, for example (density = 2·87), in the form of a thin sheet of such thickness that it weighs 1·50 mg. per sq. cm., has an air equivalent of 1·0 cm. (at 760 mm. and 15° C.). If we extrapolate for thicknesses which would correspond to the whole range in air, the above determined air equivalent does not always give the correct range of the α-rays in the substance concerned. This is due to the fact that air equivalents themselves are dependent on the velocity of the rays, and hence one and the same sheet, introduced at different points of the trajectory, shortens the range in air by different amounts.

In the investigation of the behaviour of different media with respect to the same α-rays we often take account of the 'stopping power', instead of the range or the air equivalent. The stopping power of a medium is the quantity that expresses how many atoms of a standard substance are equivalent to an atom of that medium as regards absorption. Air is usually chosen as the standard substance. What has been said of the air equivalent holds here also, but the idea of stopping power becomes unequivocal as soon as we speak of the *average* stopping power, e.g. when we understand by the stopping power of aluminium for the α-rays from RaC′ that quantity which expresses how many 'atoms' of air an aluminium atom is able to replace *on an average* throughout the whole of their path. As will be mentioned in greater detail later, the retardation of the α-rays in their passage near to or through an atom is due to the fact that the α-particle transmits energy to the atom, in which process the electrons in the atom take up orbits of higher energy content, or may be entirely removed from the atom (ionization). The stopping power must accordingly be determined primarily by the number of electrons in the atom, and secondarily by their arrangement. The dependence of the stopping power (s) on the atomic number (Z) is clearly shown in Fig. 10. It is connected with the atomic number by the relation $s = 0{\cdot}563\,\frac{Z}{\sqrt{(Z+10)}}$.

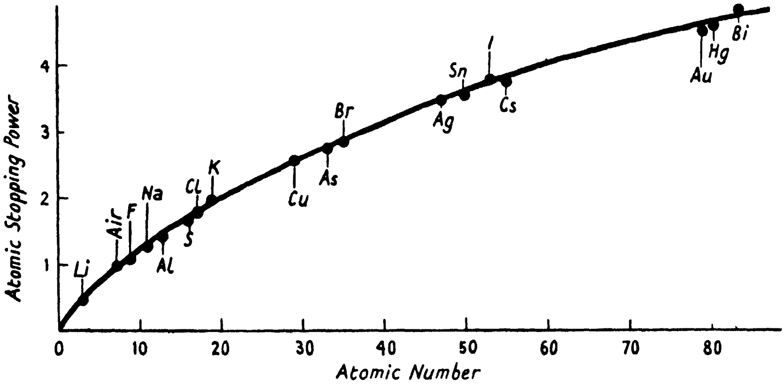

FIG. 10. Dependence of Atomic Stopping Power on Atomic Number.

The stopping power of a molecule is derived additively from those of its constituent atoms, but this relation is not strictly valid. Considerable deviations from the additive law are found only in the case of the lightest of the elements; thus from the stopping power of the H_2-molecule we obtain for the H-atom the value $s = 0{\cdot}200$, whereas the compounds C_2H_4, C_2H_6, CH_4, when we make use of the value for C found with diamond, yield the value 0·187, and from NH_3 we obtain the value 0·173. The stopping power of water vapour is smaller by 3 per cent. than that of an equivalent mixture of hydrogen and oxygen [4]. For CO_2 and CO the additive law was found to have strict validity. As applied to α-rays of different range, the stopping power of atoms shows only approximate constancy. It increases with increasing range when the atomic weight of the absorbing substance is greater than the mean atomic weight of air (14·4), and it diminishes with increasing range in the converse case. The absorption of a compounded sheet, consisting of layers of two different materials, is for this reason dependent on which side of the sheet faces the source of radiation. An exact calculation of the stopping power in terms of universal constants is possible in those cases, for which the velocity of the incident α-rays is much greater than the velocity of revolution of the electrons in the atom that are responsible for the effect. Thus it is found that calculation is in accord with experiment in the case of the lithium atom, the stopping power of which is twice that of an atom of hydrogen.

In their passage through matter, α-rays suffer a very considerable diminution in velocity, in consequence of which the phenomenon described above arises, viz. that the α-rays soon lose their characteristic properties (excitation of fluorescence, blackening of a photographic plate, ability to ionize), and so reach the end of their range. If we disregard the very last part of the track of an α-particle, a simple relation is found to exist between the velocity and the range of the particles. At any point of the track the distance remaining to be traversed is proportional to the cube of the velocity at that point (see p. 23). Thus from this relation we can calculate the initial velocity of the α-rays from RaA, which we find to be $1{\cdot}69 \times 10^9$ cm./sec; from experiments on the magnetic and electric deflexion of the rays the same value is derived. For purposes of comparison it may be mentioned that positive rays with a velocity of $2{\cdot}7 \times 10^8$ cm./sec. are able to ionize through a distance in air of about 1 mm. The velocities of the α-rays from the various radio-elements are tabulated in the third column of Table 1 (p. 22). The decrease in velocity of the α-particles from RaC′ caused by their traversing sheets of various thickness of aluminium, mica, and air is shown in Table 3.

As a rule, α-rays cannot readily be detected after their velocity has been reduced below about 0·4 of its initial value. Under particularly favourable conditions, especially in high vacua, it is possible to detect

TABLE 3

Decrease in Velocity of α-Particles from RaC′ after passing through various Materials

Relative velocity v/v_0	Mass in gm. per sq. cm.		
	Aluminium	Mica	Air
0·95	$1{\cdot}48 \times 10^{-3}$	$1{\cdot}43 \times 10^{-3}$	$1{\cdot}24 \times 10^{-3}$
0·90	2·79	2·75	2·32
0·80	5·01	4·86	4·08
0·70	7·03	6·40	5·46
0·60	8·50	7·50	6·48
0·50	9·64	8·47	7·29
0·415	10·46	9·35	7·96

α-rays down to 0·23 of their initial velocity (cf. p. 31, and Table 8, p. 33). It has also been possible to measure the diminution in velocity and in energy of an α-particle by determining the heating effect of the α-rays after they have passed through a layer of gas of variable thickness.

Accurate investigations have shown that for certain radio-elements the whole of the α-particles do not leave the atom with the same velocity. By means of deflexion experiments in very strong magnetic fields (23,000 oersteds) a fine structure of the α-particles from thorium C (range = 4·8 cm.) has been detected. The relative velocities and intensities of the individual components of this fine structure can be seen from the following Table 4 (cf. Table 24, p. 99).

TABLE 4

Fine Structure of the α-Particles from ThC

Group	Relative velocity	Relative intensity
α_2	1·0000	100
α_1	1·0034	30
α_3	0·9758	3
α_5	0·9624	2
α_4	0·9640	0·5

The difference in range between the first and second lines only amounts to about 0·5 mm. in air, that between the second and the relatively weak third line 3·5 mm., and that between the fourth and fifth lines 5·2 mm. Furthermore, both actinium C and actinon emit α-particles of different velocity. On the other hand, the α-rays emitted by the remainder of the radio-elements are homogeneous; this has been established for the rays from polonium with an accuracy of 0·3 per cent.

In the case of ThC′ and also RaC′, however, it is found that a small fraction of the α-particles have a much longer range than is stated in Table 1. Corresponding to each million α-rays emitted by ThC′ there are 65 and 180 respectively with ranges of 9·8 and 11·6 cm (see p. 23 and Fig. 9), and in the case of RaC′ there are at least nine different groups of α-particles of long range, as indicated in the accompanying Table 5.

TABLE 5

Velocity Spectrum of the α-Rays from RaC′

Velocity (in cm./sec.)	*Energy (in electron volts)*	*Relative numbers of α-particles*
(Cf. Table 1)		
$1·922 \times 10^{9}$	$7·68 \times 10^{6}$	10^{6}
1·998 „	8·30 „	0·49
2·093 „	9·12 „	16·7
2·127 „	9·41 „	0·53
2·146 „	9·58 „	0·93
2·175 „	9·84 „	0·60
2·191 „	9·99 „	0·56
2·215 „	10·21 „	1·26
2·242 „	10·47 „	0·67
2·259 „	10·62 „	0·21
		Total = c. 22 ($+10^{6}$)

In the latter case these ranges are distributed over an interval up to a range of 11·3 cm., and there are about 22 particles of abnormally long range to each million α-particles emitted by RaC′. On p. 100 will be found a discussion of the connexion between these α-particles of abnormally long range and the γ-rays that are emitted simultaneously.

The range of H-rays (see p. 67) accelerated in the forward direction by the α-rays from ThC′ is equal to 40 cm. It may be mentioned in this connexion that the range of H-rays is just as large as that of α-rays of the same velocity. Moreover, the stopping power is the same for both.

3. The Ionization produced by α-Rays

In consequence of its large velocity, an α-particle possesses at the beginning of its path a very considerable kinetic energy amounting to about 10^{-5} ergs, or roughly 10^{11} times the value of the thermal energy of a helium atom at room temperature. By virtue of this kinetic energy α-particles give rise to strong ionization along their tracks by their collisions with gas molecules. Thus an α-particle from RaC′ produces along its track of length 6·60 cm., in air at atmospheric pressure and 0° C., $2·20 \times 10^{5}$ pairs of ions, i.e. equal numbers of positive and of negative ions. The total number k of pairs of ions produced by an arbitrary α-particle amounts approximately to

$$k = k_0 R^{\frac{2}{3}},$$

where k_0 is a constant $= 6{\cdot}3 \times 10^4$, and R signifies the range at 0° C.

The intensity of ionization of the α-particles emitted by RaC′ in air, in its dependence on the distance from the source of the rays, is represented in Fig. 11. It is seen that quite near to the end of the track the ionization increases considerably, and then diminishes very rapidly. The last part BC of the graph shows a feeble tailing off, probably due to scattering. In consequence of this the range cannot be obtained from the graph with precision, so that it is customary to produce the rectilinear part AB of the graph until it cuts the abscissa, and to consider the distance of this point of intersection from the origin as the 'practical' value of the range. In the above case the maximum value of the ionization occurs 3 mm. from the end of the track of the particle. Fig. 12 illustrates the alteration which takes place in the ionizing power of α-particles as they pass through air, compared with the corresponding changes experienced by the number and by the velocity of the particles. These curves are representative of the case of the α-rays from RaC′ at 15° C.

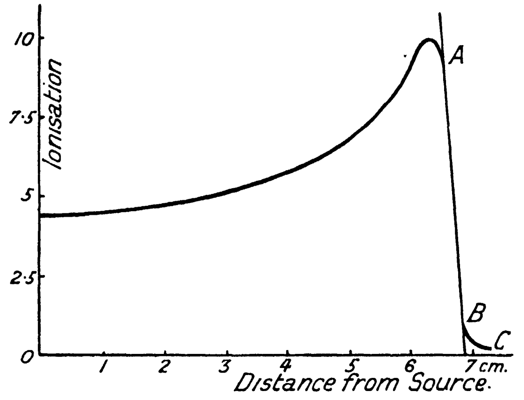

FIG. 11. Variation of the Ionization produced by an α-Particle with Increase of Distance from the Source.

If several α-rayers are simultaneously present in the source, the curve of ionization has a more complicated form. Fig. 13 shows the ionization curve of the active deposit of thorium. The occurrence of such a curve always indicates the presence of a heterogeneous radiation, or since each α-transformation is accompanied by a homogeneous radiation, it bears witness of the presence of different products emitting α-rays. Just as an analysis of the curves representing the variation of activity with time (see Chap. XI) renders possible the interpretation of the process of disintegration, so we can also make use of an analysis of the curve of ionization for the same purpose. Thus Fig. 13 shows that there are two α-rayers in the active deposit of thorium, namely ThC and ThC′, with ranges of 4·8 and 8·6 cm. respectively.

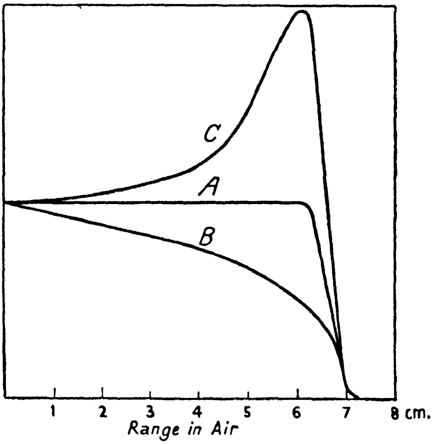

Fig. 12. Absorption of α-Rays in Air. *A*: Number of Particles; *B*: Velocity; *C*: Ionization.

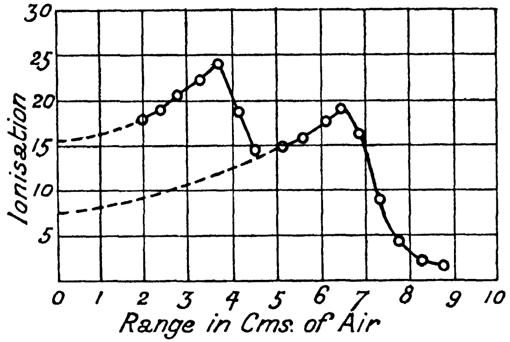

Fig. 13. Curve of Ionization due to Presence of two α-Rayers (ThC and ThC′).

When a parent substance is in equilibrium with several disintegration products, the part played by the individual disintegration products in building up the total ionization depends not only on the number, but also on the ranges of the individual α-rays. Thus the ratio of the ionization produced by ThX+Thoron+ThA+ThC to that produced by RdTh is 4·59, the relative ionizations due to these five substances being respectively 1·61, 1·78, 1·92, 1·71, and 1·53.

Since the energy necessary to produce a pair of ions depends on the nature of the gas, there exist between the values of the total ionization produced by an α-particle in various gases differences of varying magnitude, according to the velocity of the α-rays used. Thus for α-rays from polonium of residual range 7 mm. we find the following relative values for the total ionization (Table 6). The amount of energy expended by an α-particle in the production of a pair of ions is almost independent of the velocity of the α-particle. In diatomic gases the ionization is less than in monatomic gases. Most of the ions produced possess a single electronic charge. In hydrogen, oxygen, chlorine, mercury, etc., there is about one doubly charged ion to every 600 singly charged ions, whereas in helium about 16 per cent. doubly charged ions have been observed.

TABLE 6

Relative Ionization of the α-Rays from Polonium in different Gases

Air	1
Helium	1·26
Neon	1·28
Argon	1·38
Nitrogen	0·98
Oxygen	1·08

In Table 7 are shown the amounts of energy, expressed in electron-volts, which are necessary to produce a pair of ions in certain gases, together with the ionization potentials of these gases. The number of pairs of ions produced by an H-particle per cm. of its path is only one-quarter of that produced by an α-particle of the same velocity.

TABLE 7

Ionization Potentials of different Gases

Gas	*Energy necessary in electron-volts*	*Ionization potential in volts*
Hydrogen	32·5	15·8
Helium	27·6	24·5
Nitrogen	35·6	15·8
Oxygen	32·2	12·5
Argon	25·2	15·7

As already mentioned on p. 20, the counting of scintillations, in addition to ionization measurements, plays a very important part in the detection of α-particles. The scintillation method is extraordinarily useful especially when we require to detect α-particles in the presence of a strong β- or γ-radiation. Zinc sulphide screens (see p. 243) are able to transform up to 25 per cent. of the kinetic energy of α-particles into visible light energy. For a practised eye about 30 quanta of green light ($\lambda = 0{\cdot}5\,\mu$) suffice for the unaided visual perception of a scintillation.†

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† When a microscope is used for counting, the energy entering the eye must be about 300 quanta, or about 1200 quanta for reliable counting.

Using the α-rays from RaC′, the smallest velocity which is just able to give rise to scintillations amounts to 13 per cent. of the initial velocity of the rays. β-rays excite strong fluorescence in zinc sulphide, but they do not produce scintillations. If, however, several β-rays strike the same tiny zinc sulphide crystal simultaneously, sufficient luminous energy may be emitted to produce a visible scintillation.

4. Scattering of α-Rays

If we allow the α-particles from a point source to pass in a narrow pencil through thin metal foils, e.g. through a gold sheet of thickness 1/100 mm., and then to fall on a scintillation screen held in different angular positions with respect to the initial direction of the pencil, we find that a fraction of the α-rays is deflected through a small angle from their original direction. This scattering is explained in the following way. The α-particle is deviated through a minute angle in every individual atom through which it passes, in consequence of the electrical forces prevailing there; and when the α-particle passes successively through a large number of atoms in the gold foil, these atomic deviations are superimposed to form a larger and measurable deviation.

The 'scattering coefficient' ϕ, i.e. the most probable angle of deflexion for a sheet of such thickness that its air-equivalent is 1 cm., is dependent on the material of the foil, and for the α-rays from RaC′ it has the following values:

	Au	Ag	Cu	Al
$\phi =$	2·1°	1·5°	1·1°	0·6°

Since experiment has shown that the deflexion caused by an atom is roughly proportional to the weight of the atom, it follows that the phenomenon of scattering is detectable mainly in the passage of the rays through heavy atoms. ϕ increases, moreover, as the velocity of the α-particle diminishes, and is inversely proportional to the second or third power of the velocity. The deviation also increases proportionately with the square root of the thickness of the scattering material, and the same holds for its dependence on the number of atoms per c.c. of the scattering material.

As well as this 'multiple scattering' there is another very important kind of scattering, namely 'single scattering', which arises from the interaction between α-particles and atomic nuclei (see p. 95). In the above-mentioned observation of scattering by the scintillation method it was found that a few particles are scattered through relatively large angles, and quite a small number that have been deflected through 90° and more can be seen on the luminescent screen. Thus it was found that when RaC′ was used as source, one particle in 8,000 was deflected through more than 90° in the passage of the rays through thin platinum foil. These large deviations are the result of 'single scattering', for they can only originate from a single collision. The probability is vanishingly small that the α-particle in its passage through a thin metal sheet will come twice or more often so effectively close to an atomic nucleus.

If the scattering of α-rays by light atoms is observed in an intermediate angular range, multiple and single scattering may be equally important. We then speak of 'plural' scattering. The relative magnitude of multiple scattering as compared with single scattering depends on the thickness of the scattering sheet.

The scattering of α-particles can also be established by photographic means. For this purpose the source of the rays is laid on the photographic film, and after development the tracks of the particles are seen as a series of radial lines of blackened points (i.e. silver bromide particles that have been struck by α-particles), and some of these show sudden bends, sometimes through large angles. A special photographic emulsion has recently been prepared, and this facilitates the detection of the presence of a very weak α-radiation by exposure of the plate to the radiation for some months [5]. The range of the particles can be determined in this way. In experiments with the cloud method the phenomenon of scattering is very clearly revealed. Fig. 14 (Plate I) shows not only single deflexions but also the effect of a violent collision between an α-particle and an oxygen nucleus. Here the α-particle was deflected from its original path through 76° 6′, and the direction of the recoil track of the oxygen atom struck makes an angle of 45° 12′ with the initial direction of the α-particle. The photograph was obtained by taking two exposures simultaneously on the same film, so as to get the track of the rays from two directions mutually perpendicular. Scattering played an important part in the investigation of the constitution of atoms, and we shall discuss it further in that connexion. At this juncture it will suffice to mention that, from the ratio of the number of α-particles that are deflected through an angle ϕ from the original direction of the rays to the number falling on an equal area in the same time when no scattering sheet intervenes, it is possible to calculate the nuclear charge of the scattering element. The mean value of various observations in the case of platinum was 77·4, whereas the correct value is 78 (see p. 86); the corresponding figures for silver are 46·3 and 47, and for copper 29·3 and 29.

5. Capture and Loss of Electrons by α-Rays

If α-particles pass through a narrow slit and strike a photographic plate, and we observe the deviation of the beam of rays under the action of a magnetic field, we obtain in general one line corresponding to the deviation of He^{++}. At the highest vacua, however, a second line is revealed, which has suffered a deviation only half as large as that of the main line, and which is produced by singly charged α-rays (He^{+}). The smaller the velocity of the α-rays, the more distinct is the He^{+} line as compared with the He^{++} line. For the very smallest velocities there is also an undeviated line, due to neutral α-particles (He^{0}).

In its passage through absorbing atoms an α-particle can accordingly take up an occasional electron, and on rare occasions it may even capture a second electron. But these electrons are soon cast off again, and in the course of the flight of an α-particle this capture and loss of electrons is repeated several thousand times. Table 8 shows the ratio of the numbers of singly and doubly charged α-particles (He^{+}/He^{++}), which is equal to the ratio of the mean free paths (λ^{+}/λ^{++}) of the particles.

TABLE 8

Capture and Loss of Electrons by the α-Particles from RaC′
Initial Velocity $V_0 = 1{\cdot}922 \times 10^9$ cm./sec.

Velocity V/V_0	*Range in cm.*	He^{+}/He^{++} *or* λ^{+}/λ^{++}
0·94	5·79	0·005
0·76	3·06	0·015
0·47	0·72	0·133
0·32	0·23	0·53
0·29	0·17	1·00
0·27	0·14	1·5
0·23	0·08	3·0

The increase in the number of He^{+}-particles very near to the end of the range is also shown in the investigation of the deviability of the α-rays in strong magnetic fields as high as 40,000 oersteds. In these experiments it is found that the curvature of the track caused by the magnetic field does not continuously increase with decreasing velocity of the rays, but that it passes through a maximum value very near the end of the range. This result is explained by the fact that towards the end of the range He^{+}- and He^{0}-particles occur, and in consequence the mean value of the charge of the α-particles diminishes. Since a diminution in charge acts on the deviability of the rays in the contrary sense to the diminution in velocity, the possibility arises of the occurrence of a maximum curvature for a particular velocity.

The effect of the change in the charge of the α-particles is also noticeable in the occurrence of fluctuations in the range of the particles. At the moment of their emission the α-rays from a single radioactive substance all possess the same velocity (cf., however, p. 26). As soon as they have traversed a sheet of an absorbing substance, however, sufficient to reduce appreciably their velocity, the pencil of rays begins to be inhomogeneous. If we determine the range by scintillation counts, we find that the limit at which the scintillations disappear is not sharp, but that the number of scintillations gradually falls off within a region of from 3 to 4 mm. As measured by means of a point counter, the mean breadth of this region of straggling has been found to be 2·95 mm. for the α-particles from ThC′ in air at 15° C. and 760 mm. pressure. The breadth of straggling of from 3 to 4 mm. is but little dependent on the nature of the absorbing material, and is approximately just as large for α-rays of long as for those of short range. The fluctuations in range of the α-particles, at the end of their path, are related fundamentally to the fact that both the transfer of energy during individual collisions and the fluctuations in density of the gas molecules have a statistical distribution.

6. Probability Fluctuations in the Emission of α-Rays

If we count the number of scintillations produced on a scintillation screen by a constant and homogeneous source of radiation in intervals

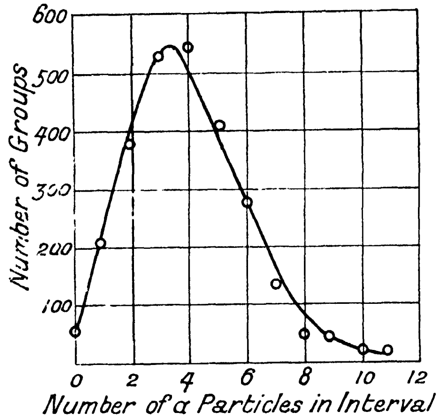

FIG. 15. Fluctuations in the Rate of Emission of α-Particles.

of, say, 15 sec. duration, we find that the values fluctuate; in some cases they are larger, in others smaller than the average value. By the average value we mean the number which results when the number of scintillations counted in, say, 2 hours is divided by 480, i.e. the number of 15-second intervals in 2 hours. The smaller the intervals during which counts are made, the greater are the fluctuations obtained. This phenomenon is a particularly simple example of probability fluctuations, the presence of which is always demanded by the statistical interpretation of natural phenomena, when the magnitudes we observe are only average values.

The abscissae of Fig. 15 show the numbers of α-particles counted in 15-second intervals, whereas the ordinates represent the number of experiments of that duration in which these numbers of particles were recorded. The results are in accord with those found from the calculus of probabilities.†

As in the emission of α-particles, we can also detect a probability fluctuation in the measurements of their ranges.

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† If a represents the mean value, then the probability p that n particles will be emitted in a given interval of time is given by

$$p = \frac{a^n}{n!}e^{-a}.$$

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III

THE β-RAYS

1. Mass and Charge of the β-Rays

The deflexions experienced by β-rays when they pass between the poles of a magnet or between the plates of a charged condenser show a great similarity in direction and in magnitude to those found with cathode rays. It has been shown by deflexion experiments in a magnetic field that, just as the cathode rays possess different velocities according to the manner of their production,† so also we find rays of different velocity in the investigation of the β-rays from radioactive substances. Here, moreover, we meet with velocities‡ of magnitude much greater than are ever reached by cathode rays; thus with RaC there are β-rays with a velocity 0·998 of the velocity of light. In accordance with the theory of relativity the mass of a body is dependent on its velocity of translation. In practice this dependence does not become manifest until the velocity is very considerable, i.e. of the order of magnitude of the velocity of light.|| Thus, in the case of β-rays of large velocity, we are led to expect a marked dependence of the mass of a β-particle on its velocity, and this has been established, in complete accord with the demands of the theory of relativity. Correspondingly, we differentiate between the 'rest mass' and the 'translational mass' of β-particles, i.e. the mass of the particle in motion. For the former the

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† When they leave the cathode the electrons already have a mean thermal energy of $1{\cdot}725\times10^{-4}\,T$ electron-volts.

‡ Apart from the velocity of an electron (v) measured in cm./sec., use is frequently made of the ratio (v/c) of the electronic velocity to the velocity of light as a measure of the velocity. In calculations, especially for small velocities, we frequently have recourse to the velocity expressed in volts (V), i.e. that difference of potential in volts through which an electron originally at rest would have to fall *in vacuo* in order to attain the velocity v:

$$V = 511{,}000\left\{\left(1-\frac{v^2}{c^2}\right)^{-\frac{1}{2}}-1\right\}.$$

A further measure of the velocity which is often used when the velocity is measured by the magnetic-deflexion method is the product of the magnetic field strength (H) and the radius of curvature (r):

$$Hr = 1{,}702\frac{v/c}{\sqrt{(1-v^2/c^2)}}.$$

|| The mass m corresponding to the velocity v is given by

$$m = m_0\left(1-\frac{v^2}{c^2}\right)^{-\frac{1}{2}},$$

where m_0 = the rest mass, c = the velocity of light. Whereas particles with a velocity 1/10 that of light only show an increase in mass $\frac{1}{2}$ per cent. greater than that of the particles at rest, the value of m for particles with 0·998 of the velocity of light is 15·8 times the value of m_0.

same value was found as for the rest mass of a cathode-ray particle, viz. $9{\cdot}105\times10^{-28}$ gm., which corresponds to 1/1,837 of the mass of an atom of hydrogen. Thus we conclude that β-rays consist of rapidly moving electrons.

When β-rays fall on a positively charged electrometer they discharge it; conversely, a preparation emitting β-rays becomes positively charged when it is insulated *in vacuo*, as can be shown by connecting it to an electrometer. Using radium preparations, and exercising great care as to the insulation, it is possible in this manner to attain a charge corresponding to a potential of +150,000 volts (see also the 'radium clock', p. 261). It is possible to determine the number of β-particles emitted by a known quantity of radium, by noting the charge acquired by an electrometer to which it is connected, in consequence of the escape of β-particles. Another method of counting β-particles is based on the use of the point counter (p. 17), which is also used with advantage for the counting of α-particles, as mentioned on p. 21.

In the case of α-transformations we saw that one atom emits only one α-particle, and it is likewise found that in the transformation of a β-rayer such as RaB or RaC each disintegrating atom emits only one β-particle. From the combined amounts of RaB and RaC in equilibrium with 1 gm. of radium $(2\times3{\cdot}7)\times10^{10}$ β-particles are emitted in each second (cf. p. 21). The simplicity of the conditions prevailing in such experiments is very much masked by the occurrence of secondary rays, and we shall discuss this when we are dealing with secondary rays.

2. Absorption and Velocity of β-Rays

Corresponding to their large velocity, the penetrating power of most β-rays is considerably greater than that of α-rays. In their passage through matter, the intensity of the rays gradually diminishes, and the thickness of the layer by which they are 'completely' absorbed depends in no small degree on the delicacy of the absorption measurements, in addition to the hardness of the rays.

If in the investigation of the β-radiation from RaE, for instance, we plot as abscissa the thickness of the absorbing sheet of aluminium, and as ordinate the ionization produced by the radiation, we obtain a simple regular graph. The values of the ordinates diminish in geometrical progression as the abscissae increase in arithmetical progression.† The intensity of the radiation is reduced to half its value in its passage through a layer of thickness $0{\cdot}16$ mm. Al, after passing through $2\times0{\cdot}16$ mm. it is reduced to a quarter, and so on. Accordingly, the 'half-value thickness' of aluminium for the β-rays from RaE has the

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† The decrease in the radioactivity of every simple disintegrating radioactive substance takes place according to the same type of curve (see graph B in Fig. 41, p. 128).

value 0·16 mm. The steeper the downward slope of the above-mentioned absorption curve, the more easily are the rays absorbed, and the smaller is the value of the half-value thickness. Instead of the last-named quantity we often consider the 'absorption coefficient', usually designated by μ,† and equal to $\frac{0{\cdot}693}{\text{half-value thickness}}$.

The absorption curves do not always show the simple form just described, which can be represented mathematically by means of a single absorption coefficient; for their interpretation it is often necessary to assume the existence of a mixture of rays, i.e. different absorption coefficients. Thus the β-rays from RaB exhibit the absorption coefficients 13, 77, and 890 when they are passed through aluminium. The absorption coefficients of different β-rays in aluminium are given in Table 9.

TABLE 9

Absorption Coefficients of different β-Rays

Radio-element	*Absorption coefficient μ in aluminium*
UX_1	460
UX_2	18
Ra	312
RaB	13, 77, 890
RaC+RaC″	13, 50
RaD	5,500
RaE	43

When the absorption coefficient is small, the half-value thickness is large, i.e. the penetrating power is large; for this reason such rays are also called 'hard', in contradistinction to the easily absorbed 'soft' rays.

If we investigate the absorption of one and the same β-radiation in different materials, we find that the absorption coefficient increases with the density of the absorber, but that the ratio of the two quantities is not strictly constant. Table 10 contains a number of values of the ratio of the absorption coefficient to the density, or the mass-absorption coefficient, for a variety of solid elements, and for the β-rays from UX_2, whereas Table 11 gives a comparison of the absorption coefficients of β-rays of different hardness in air and in carbon dioxide.

The absorbing power of a chemical compound is built up additively

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† If I_0 is the intensity before absorption, and I the intensity after the rays have passed through a layer of thickness d, then $I = I_0 e^{-\mu d}$, where μ = the absorption coefficient. Putting $I = \frac{I_0}{2}$, we find $\mu = \frac{\log_e 2}{d_{\frac{1}{2}}} = \frac{0{\cdot}693}{d_{\frac{1}{2}}}$ cm.$^{-1}$ Thus the 'half-value thickness' is related to the 'absorption coefficient' in the same way as the 'half-value period' to the 'disintegration constant'. (Cf. p. 127.)

TABLE 10

Ratio of the Absorption Coefficient to the Density of the Absorber

Substance	*Density*	*Absorption coefficient* / *density*
Carbon . . .	1·8	4·4
Sulphur . .	2·0	6·6
Boron . . .	2·4	4·65
Aluminium . .	2·7	5·26
Barium. . .	3·8	8·8
Iodine . . .	4·9	10·8
Tin . . .	7·3	9·46
Copper . . .	8·9	6·8
Lead . . .	11·3	10·8
Gold . . .	19·3	9·5

TABLE 11

Absorption Coefficients of different β-Rays in Air and in Carbon Dioxide (at 1 atmos. and 22° C.), arranged in decreasing order of hardness of the β-Rays

Radio-element	*Air*	*Carbon dioxide*
ThC+ThC″ .	0·0068	0·0129
RaE . .	0·0152	0·0297
ThB . .	0·090	0·142
UX_1+UX_2 .	0·12	0·23
RaD . .	0·64	1·69

from those of its elementary constituents, but in the case of hydrogen compounds there exist deviations from this rule.

3. Range of the β-Rays. β-Ray Spectra

It might be expected that in the matter of absorption β-rays would show a similar behaviour to α-rays. It is found, however, that β-rays emitted by the various radio-elements are absorbed in their passage through matter according to the same laws as are rays of light (cf. p. 37). This behaviour is accounted for by the fact that the β-rays emitted by a radio-element do not possess a definite velocity like the α-rays, but show a continuous velocity spectrum, which is discussed on p. 104. With the aid of a magnetic field the continuous spectrum can be analysed into its component velocities, and in this way the intensity of the individual components can be evaluated. The energy distribution in the continuous β-ray spectrum of a few radio-elements can be seen from Fig. 16 (cf. also the discussion on p. 105). The analysis of β-rays in a magnetic field yields the further result that a line spectrum of velocities is found to be superimposed upon the continuous spectrum.

It may be mentioned in advance that the continuous spectrum of β-rays originates in atomic nuclei, whereas the line spectrum is produced in the electronic shells of the atoms. These questions are discussed in greater detail on p. 101. Each line in the magnetic line spectrum of β-rays (cf. Fig. 17, Plate I) corresponds to a homogeneous radiation, i.e. to rays of definite velocity. When we investigate the absorption of such a β-radiation in aluminium we obtain graphs that are almost straight lines, at least in the earlier stages of absorption. Fig. 18 shows the absorption of β-rays having a velocity of 0·83 that of light. In order to obtain a magnitude analogous to the range of α-rays, we may produce the line *AB*. The distance from the origin to *D* then represents the 'practical' range of the β-rays, i.e. the thickness of the layer of aluminium sufficient to absorb practically all the β-rays. The approximate rectilinear form of this graph is the result of two superimposed processes, the process of scattering and the diminution of the velocity of the rays. (For details, see p. 47.) On replacing the aluminium by paper, which has a smaller scattering power, the graph assumes a somewhat concave form, whereas lead, which has a larger scattering power, gives a convex form to the graph, relative to the origin.

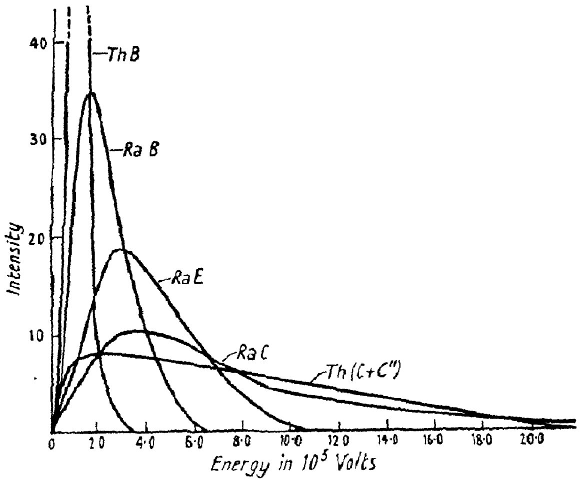

FIG. 16. Energy Distribution in the Continuous β-Ray Spectrum of Various Radio-elements.

Fig. 19 shows the 'practical' range of β-rays of different velocity in aluminium. Along the abscissa axis are plotted the velocities—in fractions of the velocity of light—and the ordinates represent the range, measured in gm. of aluminium per sq. cm.[†] The product of the range

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[†] In the case of aluminium with a specific gravity of 2·7, 1 gm. per sq. cm. corresponds to a thickness (range) of 3·7 mm.

PLATE II

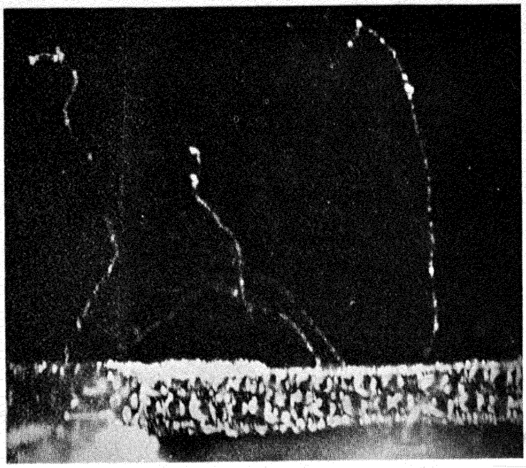

FIG. 20 (p. 41). Tracks of β-Rays from RaD in Air at 20 cm. Pressure.

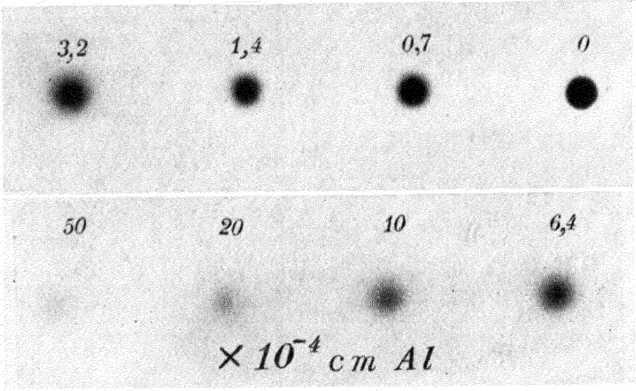

FIG. 21 (p. 45). Scattering of β-Rays after passing through Aluminium Foil.

and the density of the absorbing material is practically independent of the material. The stopping power of an atom is roughly proportional to the atomic number, but more accurate investigation reveals a periodicity of the stopping power. The presence of a well-defined range of the β-rays can also be shown by cloud photographs. Thus the β-rays from RaD comprise two strong β-ray groups having velocities corresponding to 34 and 39 per cent. of the velocity of light, and having energies corresponding to 31,000 and 43,000 volts respectively. The cloud photograph (Fig. 20, Plate II) shows two mean ranges of 0·8 and 1·58 cm. at normal pressure and room temperature. We are here dealing with 'true' ranges, or the real lengths of path traversed by the electrons before they are completely stopped. The curves that represent the dependence of the logarithm of the ionization current on the thickness of material traversed, or the logarithmic absorption curves for β-rays, are generally straight lines. Because of this fact it was at one time concluded that we were here concerned with homogeneous rays. That this is not true follows from what has been said above, and also from the fact that if, instead of deriving the absorption curve by measuring the ionization current with the aid of an electroscope, we make use of a β-ray counter (see p. 37), the resulting curve is no longer strictly exponential in form. In the interpretation of this result, however, we must take into consideration that the counter indicates a smaller fraction of the scattered electrons than the ionization method, and that the greater efficiency of the slower particles in producing ionization is not effective in the case of the counter.

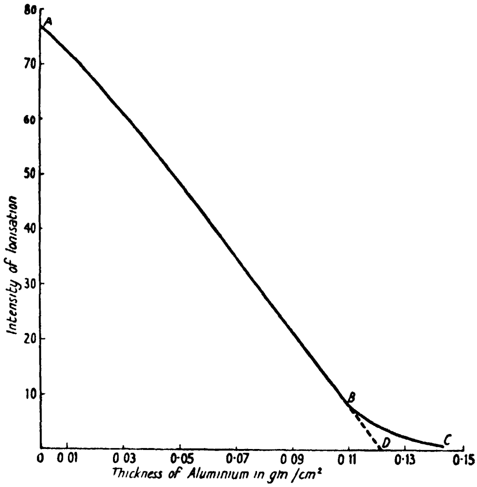

Fig. 18. Absorption of Homogeneous β-Rays in Aluminium. ($v/c = 0{\cdot}831$).

The investigation of magnetic spectra resulted in the detection of a large manifold of lines, of which, in the spectrum of RaC for example, the existence of 63 has been established. (See also p. 101.) The fastest β-rays (0·998 of the velocity of light) were detected in the magnetic spectrum of RaC, whereas in the spectrum of ThC indications are found of the existence, along with other lines, of β-rays of velocity only 0·29 of the velocity of light. In their passage through matter β-particles suffer a considerable diminution in velocity, this amounting to 30 per cent. when rays of velocity 0·95 that of light pass through 2 mm. of aluminium. In consequence of the diminution in velocity, the value of the coefficient of absorption also alters during the passage of the rays through matter. The thickness of the half-value layer is more than ten times smaller for rays of two-thirds the velocity of light than for those which possess a velocity 95 per cent. of that of light. The difference of the squares of the energy before and after penetration of an absorbing layer is roughly proportional to the thickness of the layer.

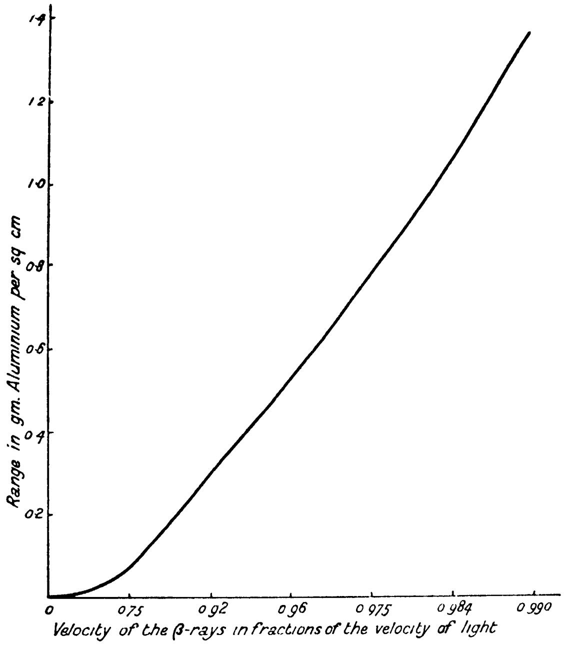

FIG. 19. Range of various β-Rays in Aluminium.

The decrease in velocity experienced by β- and α-rays when they pass through matter is essentially connected with the characteristic frequency of the electrons in the atoms, in a similar manner to the phenomena of refraction and dispersion.

4. The Ionization produced by β-Rays

β-particles ionize the gas molecules lying in their track when their velocity is not less than a certain minimum value, which in air amounts to 0·0071 of the velocity of light; for in order to ionize, the particles must have a certain minimum amount of energy. We can also consider with advantage the energy instead of the initial velocity of the β-particles, and since the energy of an electrically charged particle is equal to the product of its charge and potential (eV), and the charge of a β-particle is always constant and equal to the elementary electronic charge, we may also introduce the potential difference (V) in place of the energy to which it corresponds.† In these units the above-mentioned minimum velocity corresponds to a potential difference of 13 volts. If we endeavour to ionize air by means of cathode rays, to which we impart the necessary energy by accelerating them in a potential drop of sufficient magnitude, we find that a potential difference of 11 volts is necessary in order to obtain ionization of the air. This quantity is known as the ionization potential‡ of air. That of hydrogen has the value 13·5 volts, and that of helium 25 volts.

In spite of their large velocity, the ionization produced in a given distance by β-rays is much weaker than that effected by α-rays. A β-particle with a velocity of about 0·9 that of light produces in air at atmospheric pressure 55 positive and an equal number of negative ions per cm. of its path, or only about 1/200 the number produced by an α-particle. Since a β-particle collides with 8,400 atoms in each cm. of its path in a gas at atmospheric pressure and normal temperature, we are led to the conclusion that only about 1 per cent. of the atoms struck are ionized. The large velocity of the β-rays influences very unfavourably the yield in ions. As the velocity of the rays decreases, the number of ions produced increases considerably, but below a velocity of 0·024 that of light, at which velocity 7,700 ions are produced per cm., it begins to diminish. Corresponding to complete absorption, a β-particle of energy represented by 300,000 to 350,000 volts produces in air about 10,000 pairs of ions; the mean value of the number of pairs of ions produced per 1 cm. of path is about 70 at normal pressure. For β-rays of 39 per cent of the velocity of light (43,000 volts) it has been possible to establish by means of the cloud method the formation of 130 primary ions per cm. of the path of the particle. If we take into

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† The physical meaning of this difference of potential is that the β-particle would have acquired the same kinetic energy if it had traversed a difference of potential of this magnitude, and without having an initial velocity. (Cf. the footnote on p. 36.)

‡ The ionization potential can also be defined as the potential with which we have to multiply the elementary charge in order to arrive at the work that must be performed to remove an electron from its normal orbit within the atom to an infinite distance. (See p. 82.)

account that the secondary ionization amounts to about four times the primary ionization, the total number of ions produced by such a β-particle along the whole of its range in air can be calculated to be about 1,000. The energy expended in the production of a pair of ions corresponds to about 32 volts. Since the ionization potential of the nitrogen molecule is only about 16 volts, we must conclude from this that a considerable part of the energy handed over by a quickly moving electron in its collision with molecules is utilized in performing work other than ionization, as in the excitation of molecules and similar processes.

For velocities not exceeding about 40 per cent. of the velocity of light, the diminution in the velocity of electrons effected by small thicknesses of matter is inversely proportional to the cube of the velocity. In the case of platinum the constant of proportionality is about four times as large as with aluminium. For very large velocities (85 to 95 per cent. of the velocity of light) the decrease in the energy of the electron is proportional to the thickness of the layer of matter traversed.

The number of ions produced per 1 cm. of path in one and the same gas is proportional to the density of the gas, so long as the gas is attenuated; for different gases the proportionality between the number of ions produced and the density is only approximately valid. The above numbers have reference to the total ionization caused by β-rays. In general, the primary ionization amounts only to a fraction of the total ionization, and it arises by virtue of the fact that the primary β-particles ionize individual atoms. The number of these initially ionized atoms can be evaluated by means of the cloud method, for a water droplet is associated with each ion. The evaluation of the number of droplets along the path of a β-particle of one-third the velocity of light yields for the number of primary pairs of ions produced about 90 per cm. at atmospheric pressure. This number is roughly inversely proportional to the square of the velocity of the particles.

5. Scattering of β-Rays

Whereas the scattering experienced by α-rays in their passage through matter is detectable only by accurate observations, and is in practice without influence in absorption measurements, scattering is very important in the case of β-rays, and it is not permissible to neglect it. Thus the ionization produced in an electroscope by β-rays that have passed through an absorbing layer of, say, aluminium foil is considerably greater when the foil is situated in the immediate vicinity of the electroscope, than when it is immediately in front of the radioactive source, which is usually placed at a distance of several centimetres from the electroscope. In both cases a fraction of the rays is scattered in traversing the aluminium foil, but whereas in the former case almost the whole of the scattered radiation also enters the electroscope, in the latter it does not.

The exceedingly intense scattering experienced by β-rays even when they pass through quite thin sheets of aluminium is shown very clearly in the photographs reproduced in Fig. 21 (Plate II).

These photographs were obtained by allowing the β-rays emitted by a few millicuries of radon to pass through a small circular opening, and then to fall on a photographic film wrapped in black paper. When the experimental vessel was evacuated during the exposure, the pencil of rays was registered on the developed film as a sharply defined circular patch of blackening. The introduction of a sheet of aluminium of only $0{\cdot}7 \times 10^{-4}$ cm. thickness in the path of the rays was sufficient, as seen in the figure, to produce a distinct scattering of the rays, i.e. a diminution in sharpness of the edge of the area of blackening. For aluminium foil of thickness 50×10^{-4} cm. the scattering was already so intense that the central spot is almost invisible. Of course the total intensity of the rays remained practically unaltered, but in the latter case it was distributed over so large an area that the blackening superimposed on that resulting from the γ-rays vanished.

For the purpose of determining the magnitude of the scattering caused by different substances, we first measure the ionization I produced in a β-ray electroscope by a β-rayer situated on a supporting plate, the thickness of which is as small as possible. The resulting ionization is produced exclusively by particles that enter the electroscope directly. If we now place a sheet of the substance under investigation immediately beneath the radioactive source, some of the rays emitted in the downward direction, which do not under normal circumstances reach the electroscope, are now deflected upwards by the sheet in consequence of scattering, and the total resulting ionization I' in the electroscope is greater than I. The 'percentage reflection' from the sheet is given by $100\,\frac{I'-I}{I}$; it increases considerably with the atomic weight of the reflector, as can be seen from Table 12. The data have reference to the β-rays emitted by RaE, and correspond to thick sheets of the reflectors.

The number of particles reflected increases with increasing thickness of the sheet, from which it follows that the reflection does not take place at the surface of the reflecting substance as in the case of light rays, but in such a way that the β-rays, in penetrating the atoms of the reflector, are often so strongly deflected from their path that this deflexion bears a resemblance to reflection. The deviation of the β-rays is a consequence of the electrical forces existing within the atoms, and we shall return to this subject on p. 79. The particles reflected in this manner now pass through the atoms of the reflector in the reverse direction, and so reach the electroscope. The increase in the number of the reflected particles ceases when the thickness of the reflecting sheet is sufficient completely to absorb the reflected β-particles in their return journey through it.

TABLE 12

Reflection of the β-Rays from RaE from different Substances

Substance	*Atomic weight*	*Percentage reflection*
Bi . . .	209·0	70·9
Pb . . .	207·2	70·2
Au . . .	197·2	67·8
Ag . . .	107·9	57·4
Cu . . .	63·6	44·7
S . . .	32·1	32·1
Al . . .	27·1	30·0
C . . .	12·0	17·1

For thin layers it is found that the mean angle of deflexion of β-particles after passing through a layer of thickness x is proportional to the square root of x. For very small thicknesses the scattering is less than would be required by the square-root relation. If we compare the scattering in different elements for very small thicknesses, we find a greater scattering for elements of large atomic weight than would result if it were proportional to the atomic number. The most probable angle of scattering is inversely proportional to the energy of the rays, but this relation has not yet been firmly established for high velocities.

Every β-particle in its passage through atoms suffers as a rule an alteration both in its direction and in its velocity, energy being given up in the process to the atoms traversed. The diminution in velocity is brought about mainly by the electrons of the atom, and the change in direction by the influence of the nuclei. The electrons in the atom are thus chiefly responsible for the absorption of the β-rays, and the atomic nuclei for the scattering. As with α-rays (see p. 31), we differentiate here also between 'single', 'multiple', and 'plural' scattering. With very thin sheets the larger deviations arise as a result of a single elementary process. There is then a very small probability that a particle will suffer more than one elementary deflexion of the order of magnitude under consideration; simple or single scattering results. The processes of simple scattering can be rendered visible by the cloud method of investigation as sudden bends in the tracks of electrons (Fig. 20, Plate II). The less probable the single deflexion we observe, i.e. the greater the deflexions on which we focus our attention, the greater will be the thickness of the sheet up to which we encounter true simple scattering. The reverse holds for true compound or multiple scattering. This can only be expected for small angles of scattering and large thicknesses of the scattering material. The directional distribution of the scattered electrons can be approximately reproduced by means of Gauss's law of errors. If the total deviation is composed in general of more than one single deflexion, but at the same time the number of single deflexions is not large enough for the strict application of Gauss's distribution law, we then speak of the process as one of plural scattering.

When radiation falls perpendicularly on a sheet of material we naturally inquire as to the amounts of it which leave the sheet respectively on the emergence and on the incidence sides. For α-rays the answer is that practically the whole of the radiation leaves the sheet on the emergence side. With β-rays, on the other hand, an appreciable fraction of the β-particles is scattered in the backward direction; in fact, even before the velocity has diminished considerably with increasing thickness of the sheet, the number of electrons emitted on the emergence side already falls off because of this process of backward scattering. There occurs what we might call a backward diffusion of the rays. With the help of similar considerations to those indicated in the case of α-rays (p. 32), we can calculate the magnitude of the nuclear charge of the atoms responsible for the deviation, from the magnitude of the individual deflexions of β-rays. The evaluation of the fraction of the particles deflected through more than 90° yielded the value 8·1 for the nuclear charge of the oxygen atom. The absorption coefficient (see p. 37), or more correctly the 'coefficient of weakening', can be resolved into two components, the true coefficient of absorption and the coefficient of scattering. The 'atomic absorption coefficient', or the product of the true absorption coefficient and the atomic number divided by the density, is approximately constant within a period of the natural system of classification of the elements.

6. Comparison of the Absorption and Scattering of α- and β-Rays

Whereas the velocity of all the α-particles emitted by a unitary substance like RaF diminishes uniformly in their passage through matter, so that the pencil of rays remains homogeneous and the number of particles practically unchanged, the behaviour of β-rays is different. Here more and more particles are cut out by scattering during the transmission, and hence in the later parts of the path the ionization is due to only a small fraction of the particles emitted by the radiating source. The effect of this decrease in the number of the β-particles is greater than that of the increase in the ionizing action resulting from the reduction of velocity (see p. 43), and hence the ionization curves, in which the ionization is plotted as ordinate and the velocity of the rays as abscissa, show a downward trend from the beginning onwards for β-rays, in contrast to the curves for α-rays. Such a curve of ionization for β-rays, and hence also the curve of absorption, is the result of various factors, such as the decreasing number of the ionizing particles, the magnitude of the ionization produced by a β-particle which at first increases as the velocity of the particle diminishes, and the additional ionizing action of the scattered β-particles. These last include both those resulting from the action of the nuclear charges, and those that have been scattered by the electron shells of the atoms traversed. When they traverse matter, homogeneous β-rays thus become appreciably heterogeneous quite near the beginning of their path, and the exponential form of the ionization and absorption curves of these rays is only the resultant of the various contributory processes mentioned. We must further take into account that, whereas the velocities of all known α-rays are of the same order of magnitude and lie between $1{\cdot}4\times10^9$ and $2{\cdot}1\times10^9$ cm./sec.,† the β-rays embrace a much bigger range of velocities. β-rays can be detected with energies of less than 100 e.V. and of more than 2×10^6 e.V. On the other hand, if we follow out the tracks of 'single' β-particles, we obtain just as definite a range as with α-rays. For very slow electrons of velocity 25 e.V. or less the atoms of the rare gases are almost completely transparent; the mean free path of such electrons is thus abnormally large (Ramsauer effect).

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† The energies of α-particles with these limiting velocities are respectively $4{\cdot}1\times10^6$ e.V. and $9{\cdot}2\times10^6$ e.V.

REFERENCE

A detailed description of the properties of β-rays will be found in *Radiations from Radioactive Substances*, by Rutherford, Sir E., Chadwick, J., and Ellis, C. D. (Cambridge University Press, 1930).

IV

THE γ-RAYS

1. The Nature of γ-Rays

The third type of rays that we encounter in the investigation of radioactive processes is the γ-radiation. This is influenced neither by the presence of electric nor magnetic fields, and it presents a radiation similar in nature to that of light, in contrast to α- and β-rays, which consist of particles projected rectilinearly from radioactive substances. Just as the spectral resolution of X-rays has been effected with the aid of interference phenomena in crystals, the application of the same method to γ-rays has also met with partial success.†

When a cylindrical pencil of X- or γ-rays is incident on a crystal, diffracted X- or γ-rays are emitted in different directions from the parts of the crystal involved, in a similar manner to the way in which rays of light are diffracted from a diffraction grating. We can picture the production of the diffracted X-rays most clearly by regarding it as a reflection of the original rays from the lattice planes in which the atoms are situated within the crystal. If only one such crystal plane were available, the X-rays would be reflected independently of the 'colour' or wave-length. There are, however, innumerable such lattice planes in the interior of a crystal, always separated from each other by equal distances d. The more or less penetrating X-rays manage to reach many of these planes, and the radiation is reflected at all of them. These reflected rays suffer interference and annul each other, with the exception of those rays for which the following simple relation exists between their wave-length λ, the glancing angle ϕ, i.e. the angle between the incident ray and the lattice plane, and the distance d separating two lattice planes:

$$n\lambda = 2d\sin\phi.$$

In this expression n is an integer 1, 2, 3, etc., and indicates that reflection will also take place for those angles the sines of which are double, treble, etc., that of the smallest effective angle. The intensity corresponding to these successive integers, however, becomes smaller and smaller, just as in optics the spectra of the second, third, etc., orders become successively weaker. In fact, the formation of X-ray spectra just described is analogous to the resolution of light by means of a grating. The 'grating constant' is here the distance between two adjacent lattice planes in the crystal, and it is of a much smaller order of

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† The wave-length of the 'hard' γ-radiation is so small that even crystals are of no avail here as diffraction gratings.

magnitude than in optics, owing to the much smaller wave-length of the X-rays. For instance, in the case of a crystal of rock salt, d amounts to only $2{\cdot}80 \times 10^{-8}$ cm., whereas the grating space of a Rowland grating is about 10^{-4} cm. If we rotate the crystal whilst a narrow pencil of 'continuous' X-radiation is incident upon it, say from an X-ray tube, we find that for each of several definite values of the angle ϕ a homogeneous X-ray 'colour' is reflected with particular intensity, and superposed on the uniform scattered radiation which shows up as a continuous background in the spectrum. The presence of such an X-ray spectral line can be recognized either by means of a photographic plate, when a sharp line appears, or by means of an electroscope, which experiences a sudden increase in its discharge rate for each effective setting of the crystal. A point-counter can also be effectively utilized for this purpose.

If ϕ and d are known, we can obtain directly from the formula the value of the wave-length λ of the X-ray or γ-ray line in question. For example, it is possible by the method described to establish the existence of 23 separate lines in the γ-spectrum of RaB+RaC, their wave-lengths lying between 5 and 3,000 X-units, where 1 X-unit $= 10^{-11}$ cm. In the case of γ-rays of very short wave-length other methods are generally used instead of the method of crystal analysis, because the scattering then takes place within the individual atoms and not from the lattice planes. One other method that leads us more satisfactorily to our objective is based on the calculation of the wave-length of the γ-rays from the energy of the secondary β-rays produced by them (see p. 102). A third method is based on the calculation of the wave-length of the γ-rays from their absorption coefficient (see pp. 52 and 99).

Table 13 shows the approximate wave-length ranges for various types of radiation, including the γ-rays.

TABLE 13

Wave-lengths of the various Electromagnetic Waves

Type of rays	*Wave-length in cm.*
Electric waves . .	From ∞ to 8×10^{-3}
Infra-red waves . .	10^{-1} to 8×10^{-5}
Visible light waves . .	8×10^{-5} to 4×10^{-5}
Ultra-violet waves . .	4×10^{-5} to 10^{-7}
X-ray waves . .	10^{-6} to 10^{-9}
γ-ray waves . . .	10^{-8} to about 10^{-11}
Cosmic ray waves . .	down to about 10^{-14}

2. Absorption of γ-Rays

The shorter the wave-length of the γ-rays, the greater is their penetrating power through matter, or the greater their 'hardness'. If we plot the thickness of the layer traversed as abscissa, and the intensity of the radiation as ordinate, we again obtain to a first approximation an exponential curve,† as with the β-rays. But here the absorption coefficient μ is usually much smaller, or the half-value thickness $\left(=\frac{0 \cdot 693}{\mu}\right)$ is much larger. Whereas the intensity of the most penetrating β-radiation falls to half its initial value after the rays have passed through 0·5 mm. of aluminium, a sheet of lead of thickness 1·5 cm. is necessary to reduce the intensity of the hardest γ-rays to half its value.‡

With increasing density of the absorbing substance the absorption coefficient generally increases, and the ratio between absorption coefficient and density is found to be approximately constant, even for materials like gases and heavy metals whose densities are so widely different. Hence for a gas the absorption is proportional to the density. The extent of the deviations from this proportionality can be seen from Table 14, in which are given for different substances the absorption coefficient μ of the rays from RaC, and the mass absorption coefficient $\frac{\mu}{\text{density}}$. The softer γ-rays from RaC were eliminated by the introduction of a filter of lead 4 cm. thick. The substances are arranged in the order of decreasing density. The number of electrons present in the atom is of great importance in connexion with the ability of an atom to weaken the intensity of γ-rays, but even if we calculate the weakening per electron, a dependence on the atomic number is still manifest. The absorption effect of a compound is obtained by adding together those of the individual atoms present.

TABLE 14

Absorption Coefficient in Different Substances of the γ-Rays from RaC

Substance	μ	$\frac{\mu}{\text{density}}$
Hg	0·621	0·045
Pb	0·533	0·047
Cu	0·395	0·044
Fe	0·356	0·045
Sn	0·299	0·041
Zn	0·322	0·045
Al	0·126	0·047
S	0·091	0·046
Water	0·055	0·055
Air at 15° C.	$4 \cdot 64 \times 10^{-5}$	0·0378

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† Here again we have $I = I_0 e^{-\mu x}$, where I_0 is the intensity before transmission through the thickness x, and I is the intensity after transmission.

‡ For instance, in order to reduce the intensities of the α-, β-, and γ-rays from the active deposit of thorium to half their values, we require 0·003, 0·2, and 15·0 cm. of paper respectively.

Between the coefficient of absorption μ and the wave-length λ there exists a relation which is quite well represented by the equation

$$\mu = A\lambda^{2\cdot 8}.$$

When we are dealing with homogeneous rays the wave-length can be directly calculated from the coefficient of absorption by means of this equation. The value of the constant factor A depends on the nature of the absorbing substance. This relation is no longer valid for very hard γ-rays.

3. Scattering of γ-Radiation

In their passage through matter γ-rays are reduced in intensity in a variety of ways: (*a*) they produce recoil electrons, as a result of which the radiation suffers a diminution in frequency (Compton effect, see p. 57); (*b*) they lose energy by the production of positrons (p. 72; (*c*) they produce photo-electrons. A further complication of the conditions of absorption is caused by the fact that in the course of some of the above-mentioned processes γ-rays or recoil rays occur, and these are added to the radiation. Thus the positrons produced by the rays disappear with production of a homogeneous γ-radiation (see p. 72), which is added to the original radiation. Moreover, the production of characteristic X-rays accompanies the liberation of photo-electrons, and these X-rays will likewise manifest themselves in the weakened beam of radiation. When γ-radiation traverses very thick absorbing layers the occurrence of a very hard X-radiation is also observed, which was not originally present, and which arises from the production by the recoil electrons (Compton electrons) of a very hard impulse radiation. For light elements the absorption takes place primarily by virtue of the Compton process. Thus for the γ-rays from ThC″ the reduction in intensity in aluminium due to the production of positrons amounts to only 1 per cent. of that brought about by the Compton process; in the case of iron the value is 7 per cent., and for lead it is as high as 22 per cent. The absorption resulting from the liberation of photo-electrons in lead amounts to 30 per cent. of the Compton absorption in the same metal.

Whereas the Compton absorption per electron is independent of the atomic number of the absorbing element, the calculated absorption per electron due to the liberation of positrons is directly proportional to the atomic number, and the absorption due to the production of photo-electrons is roughly proportional to the fourth power of the atomic number, except in the case of the heaviest elements for which the increase is less rapid [1]. The ratio of the frequency of occurrence of a Compton scattering process and a photo-electric process increases rapidly with increasing hardness or diminishing wave-length of the rays. If the absorbing substance is uranium, then for rays of wavelength 24·4 X.U. the above ratio is almost unity, whereas for the hardest γ-rays of wave-length 5·6 X.U. the frequency of a scattering process is twenty times greater than that of a photo-electric absorption process. In light materials such as carbon the role of scattering is much greater still, for here the above-mentioned ratio has the respective values 3,000 and 80,000. In such materials, for short-wave γ-rays like the hardest component of the radiation from RaC, true (photo-electric) absorption fades into insignificance as compared with scattering. When a relatively thin filter is used in conjunction with the rays, the absorption coefficients are found to depend appreciably on the absorbing medium, this being ascribed to the absorption of the softer components of the radiation. The diminution of the full absorption coefficient or the hardening of the γ-rays from radium after transmission through lead filters of increasing thickness is shown in Table 15.

TABLE 15

Alteration of the Full Absorption Coefficient of the γ-Rays from Radium with the Thickness of the Lead Filter

cm. Pb	μ_{Al}	μ_{Pb}
0	0·176	..
0·5	0·153	..
1·0	0·144	0·715
2	0·137	0·615
2·5	0·135	..
3	0·129	0·540
4	0·126	0·533
7	..	0·533

A more thorough investigation shows that, in general, the γ-radiation yielded by the individual radio-elements is not homogeneous, but that it can be resolved into a few practically homogeneous components, i.e. into components that possess different full absorption coefficients. Table 16 shows the values of the full absorption coefficients established for different radio-elements. Thus in the disintegration of RaB, which is transformed into the bismuth isotope RaC, we recognize the emission of three different types of radiation, the softest of which has an absorption coefficient of 230 and the hardest 0·57 cm.$^{-1}$ in aluminium. If bismuth be rayed with hard X-rays, the bismuth reacts to this treatment by the emission of a kind of fluorescence radiation known as the characteristic radiation of bismuth, which consists essentially of three types of rays, the *K*-, *L*-, and *M*-radiations (see p. 63). The characteristic radiation can also be excited by the bombardment of bismuth with correspondingly energetic β-rays.† We are thus immediately confronted with the question as to whether the three types of rays emitted by RaB are identical with the characteristic radiations of bismuth, produced in the atoms meanwhile transformed into RaC, during the escape of, and by the γ-rays originating in the RaB nuclei. The truth of this interpretation has been established, and accordingly the designations *M*-, *L*-, and *K*-radiations in Table 16 signify not the characteristic X-radiations of the disintegrating atoms but those of the resulting atoms.

TABLE 16

Full Absorption Coefficients (μ) of the γ-Rays

			μ *in aluminium*				
Radio-element	*N*	*Nature of Decay*	*M-radiation*	*L-radiation*	*K-radiation*	*Nuclear radiation*	
UX$_2$	91	β	..	..	..	..	0·14
UX$_1$	90	β	..	24	0·7	..	..
MsTh$_2$	89	β	..	26	..	..	0·116
Io	90	α	1,088	22·7	0·41	..	..
RdAc	90	α	..	25	..	..	0·19
Ra	88	α	354	16·3	..	0·27	..
AcX	88	α	..	..	..	..	..
RaC	83	β	..	..	..	0·23	0·127
RaE	83	β	..	..	..	0·25	..
RaD	82	β	..	45	1·00	..	..
RaB	82	β	230	40	0·57	..	..
AcB	82	β	120	31	0·45	..	..
ThB	82	β	160	32	0·36	..	..
RaF	84	α	585	..	..	..	..
AcC″	83	β	..	..	..	..	0·198
ThC″	83	β	..	..	..	..	0·096

In various other cases, however, one or two full absorption coefficients are also found, which are to be ascribed to very hard rays emitted by the atomic nucleus itself; they cannot be interpreted as having arisen in the atom during the escape of an α- or β-particle. In accordance with this, the last column of Table 16 is designated 'nuclear radiation'. We shall return to this question in Chapter IX, after the discussion of the problem of atomic structure. The already described diminution in the value of μ with increasing filter thickness is due to the gradual cutting out of the softer rays. The truth of this is exemplified by the fact that in the case of the γ-radiation from ThC″, which is almost homogeneous even before being filtered, such a hardening effect does not take place.

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† The following relation exists between the velocity of the β-rays (electrons) expressed in kilovolts (V) and the wave-length (λ) of the hardest γ-rays (X-rays) produced:

$$\lambda = \frac{12{,}340}{V},$$

where λ is expressed in X-units (10^{-11} cm.).

4. Absorption Measurements with γ-Rays as applied to Radioactive Analysis

If we are presented with a mixture of radio-elements, the increasing reduction of the intensity of the radiation as the thickness of the absorbing material increases shows a more complicated behaviour, and such measurements can under certain circumstances serve for the qualitative detection of the presence of different radio-elements, just as in the case of the α-rays already discussed. This method can be applied to find out whether a preparation that must not be opened consists of mesothorium or of radium, and if the former is the element present, we may even establish when the preparation was made. This result is rendered possible by the fact that the penetrating γ-radiation from radium is derived exclusively from RaC (0·50 cm.$^{-1}$ in Pb), whereas from a mesothorium preparation two hard types of γ-rays are emitted by the respective γ-rayers $MsTh_2$ (0·62 cm.$^{-1}$ in Pb) and ThC″ (0·462 cm.$^{-1}$ in Pb). Now whilst equilibrium is established between $MsTh_1$ and $MsTh_2$ only a few days after the preparation is made, the ThC″ radiation gradually increases over several years, corresponding to the growth of RdTh, and hence the harder ThC″ radiation in mesothorium preparations must be more strongly represented the older the preparation is. The presence of a particularly hard radiation manifests itself by virtue of the fact that, in comparison with a definite γ-radiation such as that from a radium preparation, thick plates of lead suppress the ionization less than in the case of a softer radiation. The measurement of the ionization produced after filtration in 0·5 cm. lead is mostly used for the comparison of the intensities of the γ-radiations emitted by different radio-elements [2].

TABLE 17

Radium-Equivalent of Mesothorium and Radiothorium Preparations of Different Age

	Thickness of absorbing sheet of lead in cm.									
Preparation	*0·5*	*1·0*	*2·0*	*3·0*	*4·0*	*5·0*	*6·0*	*7·0*	*8·0*	*9·0*
MsTh, freshly prepared	1·00	1·02	0·96	0·88	0·79	0·71	0·64	0·60	0·57	0·55
MsTh, c. 3 yrs. old	1·35	1·38	1·38	1·35	1·32	1·29	1·27	1·27	1·29	1·32
MsTh, c. 9 yrs. old	0·90	0·93	0·97	1·00	1·02	1·04	1·07	1·11	1·15	1·20
RdTh	1·00	1·06	1·17	1·28	1·40	1·52	1·64	1·76	1·89	2·02

The accompanying Table 17 illustrates the behaviour, as regards absorption, found with mesothorium preparations of different ages; the measurements were performed with a preparation of mesothorium containing about 20 per cent. radium.† The values are expressed in

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† The radium content of mesothorium freshly prepared from Brazilian monazite is

terms of the ionization produced by a radium preparation, used as a standard. (In this connexion cf. also Fig. 48 on p. 233.)

From this table it is clear that when we have no knowledge of these conditions, the amount of mesothorium present as compared with radium is underestimated in the case of young preparations, especially when thick sheets of lead are used. As the age of the preparation increases, the amount of mesothorium is overestimated, and, at a later stage, again underestimated. Moreover, if radium be present in the mesothorium, it can be detected qualitatively by the corresponding alteration in time of the absorption. Nevertheless, the quantitative estimation of the different substances is fraught with great difficulty, mainly because in practice we nearly always have to deal with mixtures of mesothorium of unknown age with the element radium. Owing to their practical importance, absorption measurements with radiothorium have also been included in Table 17.

The age and the proportion of mesothorium in radium salts can also be calculated, likewise without opening the preparation, from the ratio of the respective developments of heat by the radium preparation containing mesothorium and by a standard radium preparation. (In this connexion see p. 246 et seq.)

5. Ionization produced by γ-Rays

The γ-rays do not ionize directly, but split off electrons from the molecules with which they collide, and these electrons, as β-rays, ionize the gas molecules present. That no direct ionization takes place follows, in the first place, from the fact that the penetrating power of the secondary electrons liberated by the incidence of γ-radiation on matter is independent of the length of the air-path previously traversed by the γ-rays. Had the γ-rays given up energy by direct ionization along this path, the energy of the secondary electrons would depend on the length of the air-path already traversed by the γ-rays. In the second place, we can deduce the absence of direct ionization from the appearance of cloud photographs of a beam of γ-rays. The ionization tracks shown by such photographs are always associated with the tracks of β-particles.

The γ-rays emitted by the amount of RaC in equilibrium with 1 gm. of radium produce a total of 11×10^{14} pairs of ions per sec. along their paths. This number is almost equal to the number of ions produced by the β-rays from the same quantity of RaC; but since the lengths of the tracks of γ-rays far exceed those of β-rays, it follows that the γ-rays produce a correspondingly much smaller number of ions for each cm.

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responsible for 20 to 25 per cent. of the total γ-activity, and gradually increases in relative importance in consequence of the decay of the mesothorium.

of their path. Under normal conditions the half-value thickness of air for these γ-rays amounts to 150 m., the corresponding thickness for β-rays being 0·63 m. As mentioned on pp. 37 and 43, it has been established that each disintegrating RaC atom emits one β-particle, and that this produces 54 pairs of ions along 1 cm. of its path. We can also speak of individual γ-impulses or 'photons', and calculate that a 'γ-particle', more correctly a photon, produces 1·5 pairs of ions per cm. of its path. Such a corpuscular conception of γ-rays often simplifies the solution of problems relating to γ-rays, and use is frequently made of it (see p. 111, footnote). The number of γ-photons emitted per sec. by the amount of RaB+RaC in equilibrium with 1 gm. Ra is equal to $7{\cdot}4\times 10^{10}$ (cf. pp. 21 and 37), from which we may conclude that each disintegrating atom emits approximately one γ-photon.

Since γ-rays eject β-particles from all substances, and not only from molecules of gases, the action of γ-radiation on an electroscope must depend on whether the absorbing medium, such as a lead plate, is situated near to the radiating preparation or to the electroscope, for in the former case the freed electrons or secondary β-rays from the lead will reach the electroscope in much smaller quantity than in the latter. In like manner the γ-rays liberate electrons from the whole neighbourhood of the electroscope, from the walls of the room, from tables, etc., and these produce γ-rays in their turn. This disturbing influence of the surroundings in the measurement of the absorption of γ-rays can be eliminated by completely enclosing the active source in a spherical shell of the absorbing medium. By using electroscopes or ionization chambers lined with paper or similar light substance we can minimize the disturbing action of the secondary rays formed. This will be clear from the discussion in the following chapter, a knowledge of which is essential before we can understand in its entirety the phenomenon of the scattering of γ-rays. Whereas the alteration in wave-length suffered by γ-radiation in the scattering process is independent of the wave-length,† the intensity of the Compton radiation, altered in the scattering process, depends very markedly on the wave-length. The intensity of this Compton radiation assumes considerable proportions only for rays of wave-length shorter than about 300 X.U.

In the medical application of γ- and X-rays the radiation intensity is expressed in so-called röntgen (r) units. The dose at any point is measured by the air-ionization equivalent of that part of the quantum energy which would be transformed into corpuscular energy by absorption

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† The gain in wave-length is given by $\lambda'-\lambda = 24{\cdot}2(1-\cos\theta)$ X.U., where λ = the original, and λ' = the increased wave-length, and θ = the angle between the incident and the scattered ray. The energy of the radiation produced is given by $h\nu' = h\nu-\frac{1}{2}mv^2$, where $h\nu$ is the energy of the original γ-radiation, and $\frac{1}{2}mv^2$ is the energy of the recoil electron set free by the scattering process.

in an infinitesimal volume of dry air at 0° C. and 760 mm. Hg pressure situated at that point, divided by the volume of the air, measured in cubic centimetres. If the amount of ionization is such that the total charge per c.c. on the ions of one sign would be 1 e.s.u., then the *dose* is defined as 1 röntgen (1 *r*) [3]. In terms of these units the γ-radiation from 1 mg. Ra(RaB+RaC) supplies 8·6 *r* per hour at a distance of 1 cm., whereas the unfiltered β-radiation from the same quantity supplies 1,720 *r* per hour.

REFERENCES

The properties of γ-rays are discussed in detail in *Radiations from Radioactive Substances*, by RUTHERFORD, CHADWICK, and ELLIS (Cambridge University Press, 1930), and in *Radioaktivität*, by K. W. F. KOHLRAUSCH (Akademische Verlagsgesellschaft, Leipzig, 1928).

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V

THE SECONDARY RAYS

(THE MUTUAL PRODUCTION OF THE DIFFERENT KINDS OF RAYS)

1. The Nature of the Secondary Rays

There exists an extensive interaction between corpuscular rays like the β-rays, and to a lesser extent the α-rays, and rays of an electro-magnetic nature. X-rays arise when cathode rays strike the anticathode of an X-ray tube and are brought to rest; analogously, X- or γ-rays are produced when β-rays lose energy in their passage through matter. The hard γ-rays have their origin in the nucleus of the atom (cf. p. 101). In their turn X-rays and γ-rays knock off electrons or secondary β-rays from the atoms on which they impinge, and provided they have sufficient energy, these will again liberate secondary X- or γ-rays, and so forth.

β- and γ-rays that have originated in this way are briefly designated 'secondary' rays, a term which is also applied to the rays of secondary origin produced by the α-rays. Scattered primary rays also, which are with difficulty or not at all distinguishable from secondary rays, are often counted in with the secondary rays.

The most important properties of the secondary rays will now be discussed.

2. Secondary β-Rays produced by γ-Rays

The initial velocity of the β-rays excited in matter by γ-rays is independent of the intensity of the γ-rays, and to a first approximation also of the nature of the material; thus the β-radiation produced by the γ-rays from RaC is found to possess, independently of the material in which it is excited, a full absorption coefficient in aluminium of about 20, and a corresponding half-value thickness of 0·35 mm. On the other hand, the initial velocity is dependent on the hardness of the exciting γ-radiation; the shorter the wave-length of this, the higher is the velocity of the resulting β-particles.† The number of secondary β-particles is proportional to the intensity of the absorbed γ-radiation, and increases with the atomic weight of the material used.

The secondary rays are emitted in various directions, but the direction of the primary rays preponderates, especially for substances of low atomic weight. According as to whether the secondary rays are

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† The following relation exists between the maximum energy eV of the secondary β-rays produced, and the frequency ν of the γ-rays exciting them: $U_{\text{max}} = eV = h\nu$ (cf. p. 62). In the calculation of the actual energy of the secondary β-rays (photo-electrons) it must be taken into account that, in order to release them from the atom, a certain amount of work E_s is required, so that $U = h\nu - E_s$.

emitted from the side of the sheet on which the primary rays are incident, or from which they emerge, we speak of 'incidence' or 'emergence' radiation. The intensity of the incidence radiation increases asymptotically with the thickness of the material, and reaches half its maximum value in the case of aluminium for a thickness of 0·5 mm.; it also increases with the atomic weight of the secondary radiator, as shown by the data of Table 18.

TABLE 18

Dependence of the Incidence Radiation on the Atomic Weight

Substance	*Intensity of the incidence radiation in relative units*
Lead	7
Copper	4
Iron	3·7
Aluminium	2·1
Paper	1

It is also immediately clear from the above numbers that, as already mentioned on p. 57, we can greatly minimize the influence of the secondary rays generated in the walls of the electroscope during the measurement of γ-rays, by making use of an electroscope constructed from paper. Special precautionary measures are still necessary, however, in order to reduce the secondary β- and γ-radiations from the walls of the room, from tables, and so on. The intensities of the incidence and emergence radiations are markedly different, as can be seen from Table 19, in which the data have reference to relatively thick sheets of the secondary radiators. When quite thin foils are used, the emergence radiation is more intense than the incidence radiation, even for the heavier elements.†

TABLE 19

Intensity of Secondary β-Rays

Secondary electrons from	*Soft γ-rays*		*Hard γ-rays*	
	Incidence radiation	*Emergence radiation*	*Incidence radiation*	*Emergence radiation*
C	170	2,280	58	1,150
Al	280	1,810	120	795
S	340	1,575	154	685
Fe	487	1,350	163	560
Cu	558	..	202	523
Zn	618	1,160	224	485
Sn	1,051	1,170	333	303
Pb	1,723	2,001	497	470

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† In practice we meet with a mixture of incidence and emergence radiations, the composition of which depends largely on the experimental arrangement used.

The secondary β-rays are composed partly of photo-electrons, and partly of recoil electrons (cf. p. 52). In the production of the former the whole of the energy of the γ-quantum or photon, reduced by the work of extraction (cf. footnote, p. 59), is transmitted to the electron, whereas in the production of recoil electrons only a fraction of the photonic energy is handed over to the recoil electron. For hard γ-rays the recoil electrons preponderate. With decreasing hardness of the γ-rays and increasing atomic number of the secondary rayer, however, the ratio turns the scale rapidly in favour of the photo-electrons. Using air as the secondary radiator and X-rays of hardness equal to that of the K-radiation of lead, the ionizing action of the recoil electrons only amounts to about 1 per cent. of that of the photo-electrons. Secondary electrons are also emitted from the surface of a solid body, e.g. a metal, when it is struck by electronic rays. Electrons of tertiary origin also frequently occur in this process. The number of secondary electrons corresponding to each incident primary electron depends on the velocity of the primary electron, on the angle of incidence of the primary rays, and on the nature and surface condition of the body. The gas content of the surface also has a fundamental influence on the effect. In certain cases the number of secondary electrons emitted may exceed the number of primary electrons absorbed, so that if the body is insulated, it charges up positively. If secondary electrons are measured along with the transmitted primary radiation, we may likewise obtain an apparent 'negative absorption'. The velocity of the secondary rays does not depend essentially either on the material of the secondary rayer or on the velocity of the primary rays. The tertiary electrons can be distinguished from the secondary electrons by the aid of the cloud-track method. The cloud droplets, each of which registers an ion, are arranged along the track of the rays not only in single pairs, but also in groups of 4, 6, and so on. Such a group indicates that a secondary electron has produced 1, 2, or more tertiary electrons. On the average 2 or 3 tertiary electrons result from each secondary electron, when the primary β-radiation has a velocity 33 per cent. that of light.

3. Electromagnetic Rays produced by Electrons[†]

In order to produce X-rays of wave-length λ the energy and corresponding velocity of the cathode rays must be greater the smaller the value of λ, i.e. the greater the 'hardness' of the resulting γ-rays. The greater the available energy of the cathode rays, the more considerable will be the hardness of the γ-rays. If instead of λ we consider the frequency

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[†] It has been possible to investigate the excitation of X-rays by cathode rays on a much broader basis than the excitation of γ-rays by β-particles. In view of the close analogy between the two cases and the importance of this class of phenomenon, the behaviour of X-rays is also briefly treated here.

$\nu = c/\lambda$, where c = the velocity of light, the following relation holds: $U = eV = h\nu$, where U is the energy of the cathode rays, V is the discharge potential, and h is Planck's constant.† Alternatively, we have $\lambda = 12{\cdot}34/V$, when λ is expressed in Angstrom units (1 A.U. = 10^{-8} cm.), and V in kilovolts (cf. p. 54). From this relation it follows that the shortest wave-length that can be excited by the aid of a discharge potential of 10,000 volts is of magnitude 1·234 A.U.

Whereas the validity of the above discussion is independent of the chemical nature of the radiating source, we shall discuss in what follows the influence of the nature of the source on the character of the X-radiation emitted.

If, for instance, cathode rays impinge on a rhodium anticathode in an X-ray tube, an investigation of the X-radiation emitted by it shows the presence, along with a heterogeneous 'white' radiation, of several sharply defined types of rays, of which the shortest has a wave-length of 0·534 A.U. This radiation is as 'characteristic' of rhodium as the *D*-lines in optics are characteristic of sodium.‡ As required by the above formula, it ceases to occur as soon as the discharge potential sinks below about 23,000 volts. Further lines of the rhodium spectrum, those of the *L*-group, can only be detected by the use of a vacuum spectrograph, for they are so soft that a layer of air 20 cm. in thickness almost completely absorbs them. Fig. 22 (Plate III) shows the *L*-spectrum of the mineral thucolite, and was obtained by attaching a small amount of the mineral to a copper anticathode. In it the absorption edge of bromine can also be recognized. As soon as the hardness of the rays falling on the photographic plate attains a value sufficient for the excitation of the *K*-radiation of the bromine contained in the plate, a sudden increase occurs in the absorption of the rays, and hence in the blackening of the plate. X-ray spectroscopy further enables us to detect with ease most substances, provided they are present in concentrations not less than 0·01 to 0·001 per cent. The sensitivity is smaller in the case of light atoms, since here a considerable part of the X-rays may be absorbed in the very atoms in which they are excited, giving rise to photo-electrons ('Auger Effect').

The difference between the two types of radiation, the impulse or continuous radiation and the characteristic radiation, in respect of their distribution of wave-length in the spectrum is most clearly brought out by having recourse to an optical analogy. The impulse radiation corresponds to continuous white light, whereas the characteristic radiation has its optical analogue in a line spectrum.

In the X-ray spectrum we differentiate between the *K*-, *L*-, *M*-, *N*-,

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† Planck's constant $h = 6{\cdot}61 \times 10^{-27}$ erg sec.

‡ Just like the *D*-line, the strongest rhodium line also consists of a doublet (0·614 A.U. and 0·619 A.U.).

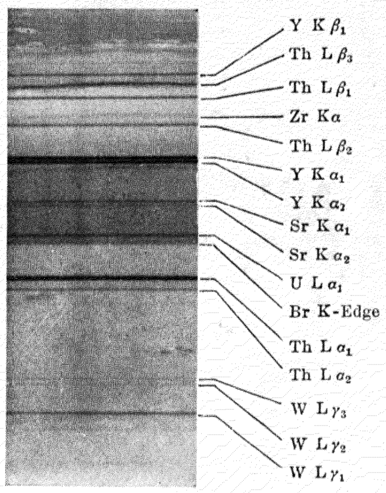

FIG. 22 (p. 62). Emission Spectrum of the Mineral Thucolite.

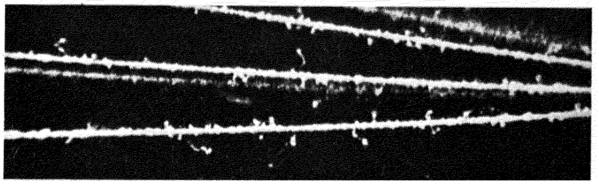

FIG. 27 (p. 67). Tracks of δ-Rays (magnified cloud-tracks of α-particles in helium at low pressure).

O-, and P-series, the wave-lengths of which increase as we pass in the same order from series to series. The relative positions of the lines in the first three series are shown diagrammatically in Fig. 23. As far as we know at present, there are at most 12 lines in the K-series, and 22 in the L-series. These lines can also be divided into different sub-groups; whereas we distinguish only one group in the K-series, we find three in the L-series, five in the M-series, seven in the N-series, five in the O-series, and three groups in the P-series. Correspondingly, as we shall see in further detail when we discuss atomic structure, we recognize in the case of the heavy elements $1K$-, $3L$-,† $5M$-, $7N$-, $5O$-, and $3P$-levels. The excitation potential of the cathode rays producing them is the same for all lines of the same group. The characteristic rays of an element are related in a simple manner to the atomic number. Thus, for the frequency or 'wave number' ν ($= 1/\lambda$) of the strongest line in the K-series, the following approximate relation, due to Moseley, holds:

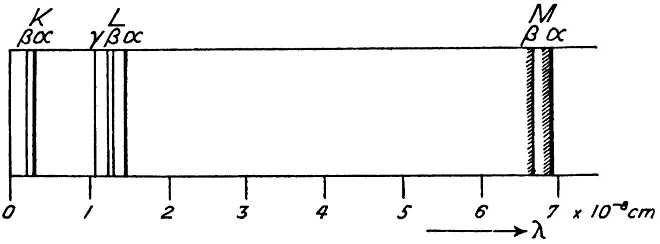

FIG. 23. Wave-lengths in the K-, L-, and M-Series.

$$\surd\nu = \surd(\tfrac{3}{4}\nu_0)(N-1),$$

where N = the atomic number (= 1 for H, 2 for He, 3 for Li, etc.), and $\nu_0 = 1{\cdot}09737\times10^5$ cm.$^{-1}$, the well-known Rydberg constant in optics.

Apart from a small systematic deviation, the following formula holds for every line in the characteristic spectrum:

$$\surd\nu = A(N-B),$$

where A and B are constants, and N is the atomic number. If we construct a graph with the atomic numbers of the elements as abscissae and the square roots of ν as ordinates, we obtain almost straight lines, as seen in Fig. 24, the data of which refer to the strongest lines of the K-, L-, and M-series respectively. We shall deal later with important conclusions which may be drawn from this regularity. Suffice it to say at this juncture that this feature in itself reveals the superiority of the

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† Beginning with the level that corresponds to the most intense absorption edge, these levels are designated L_1, L_2, and L_3 levels, frequently also L_{III}, L_{II}, and L_I levels ($L_1 = L_{III}$).

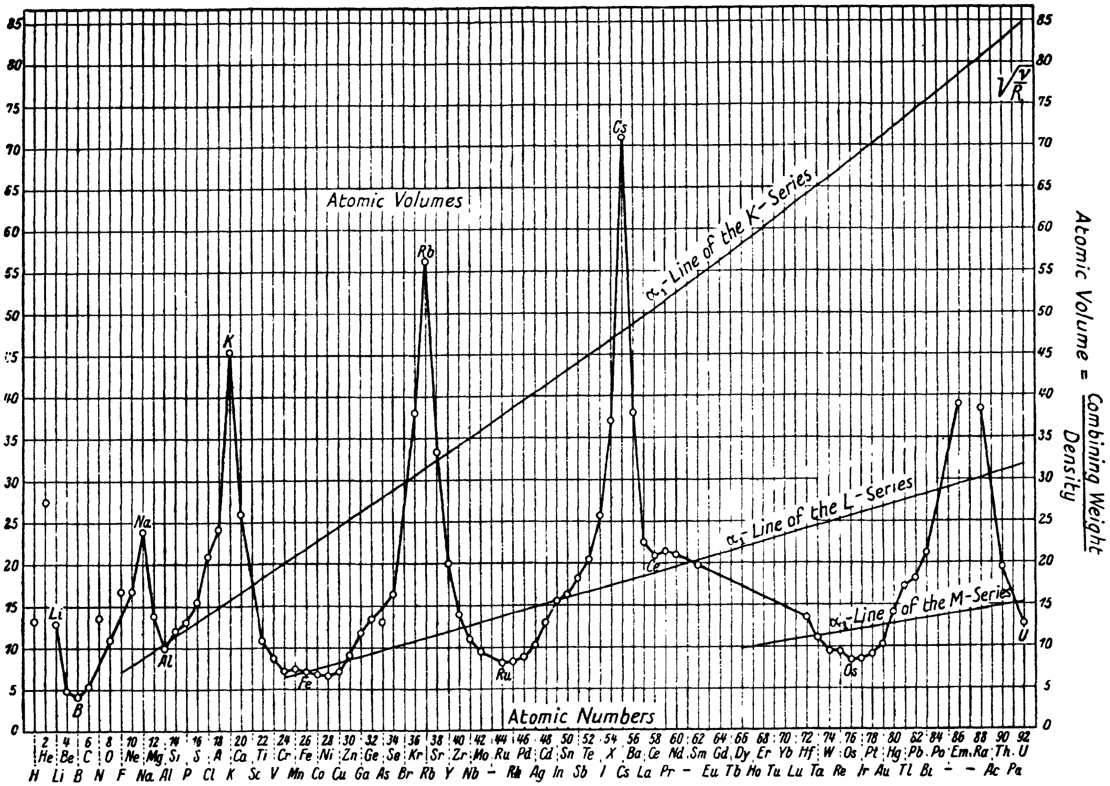

Fig. 24. X-Ray Lines and Atomic Volumes of the Elements, plotted against their Atomic Numbers.

atomic number over the atomic weight as a means of classifying the chemical elements, for if we choose the atomic weights as abscissae, we do not obtain straight lines, but irregular ones.

The production of characteristic rays results also from the action of X-rays, when their frequency is not less than that of the characteristic limiting frequency (see p. 89). It is the same condition as that which governs the occurrence of fluorescence in optics, in accordance with Stokes's law; a fluorescence radiation can be excited only by a radiation that lies farther towards the violet end of the spectrum. Since most γ-rays are of high frequency, they are able to excite the characteristic radiations of many substances in their passage through them, and this, like the scattering of the γ-rays and the occurrence of secondary β-rays, gives rise to a considerable complication of the phenomenon of absorption. It should be mentioned here that the excitation of the softer K-lines requires the same potential as is necessary for the hardest line of the K-series of the element concerned. In the calculation of the least potential necessary to excite a characteristic line, we must, in fact, introduce into the equation on p. 62 not the wave-length of the particular line but always the wave-length of the absorption edge of the group involved, i.e. of the K-group in the present case (cf. p. 63). Thus the hardest characteristic radiation known, viz. the K-radiation of uranium, has a wave-length of 101 X.U. and requires an excitation potential of 115 kilovolts.

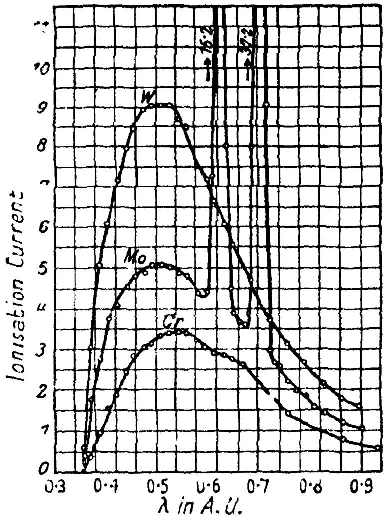

FIG. 25. Distribution of Intensity of Radiation with Wave-length.

The limit of the spectrum of the continuous or impulse radiation on the short wave-length side is determined only by the magnitude of the potential necessary for its production (see p. 62). The total energy of the radiation is proportional to the square of the applied potential, and hence to the fourth power of the velocity of the electrons producing it. Moreover, at constant potential, the total radiation is proportional to the atomic number of the element in which the X-radiation is excited. The distribution of the intensity of the radiation for different wave-lengths is shown in Fig. 25, for the region lying between 0·36 and 0·93 A.U. (360 to 930 X.U.). The ionization current, which is plotted as ordinate, serves as a measure of the intensity of the radiation. The sudden marked rise in intensity in the case of molybdenum is due to the fact that the characteristic *K*-radiation of molybdenum falls in the region investigated, whereas the much harder *K*-radiation of tungsten and the softer *K*-radiation of chromium fall outside the above range of wave-lengths. The intensity of the emission spectrum thus experiences a sudden increase in those positions where the characteristic rays of the element are excited, and in consequence the emission lines stand out prominently on the more or less blackened background of the photographic plate. In the same way a strongly enhanced absorption of the rays also takes place in definite positions depending on the position of the characteristic spectrum. If X-rays are allowed to traverse a sheet of molybdenum, it is found that strong absorption suddenly sets in at 0·618 A.U. For the case of silver, Fig. 26 shows this sudden increase in the absorption, which takes place as the wave-length is gradually reduced. The number of such absorption edges is much less than the number of emission lines, and this is due to the fact that, for the excitation of each line, it is not sufficient that the condition $eV = h\nu$ be satisfied; the energy used up in the excitation must rather suffice to excite the whole group, i.e. all the *K*-lines. Hence it is that only one absorption edge is associated with each group of lines; in consequence of this there exists only one *K*-edge, and correspondingly only one jump also in the curve for molybdenum (cf. Fig. 26) in the *K*-region. On the other hand, for the softer rays of the *L*-region there are three jumps in the curve, and these correspond to the three *L*-absorption edges, but they are no longer visible in the figure.

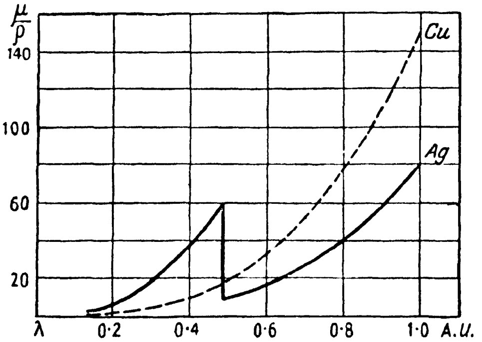

FIG. 26. Variation of Absorption Coefficient with Wave-length.

4. Secondary Rays produced by α-Rays. δ-Rays

Secondary β- as well as γ-rays may arise also when α-rays impinge on matter. If the α-rays strike solid bodies, slow electron rays or δ-rays are liberated from the place of impact. The number of δ-rays released by an α-particle is practically independent of the nature of the metal and of the direction of incidence of the α-particle. On the other hand, the number of δ-rays increases as the velocity of the α-rays decreases. The presence of gases in the material bombarded plays an important part in the emission of the δ-particles. Thus a surface of zinc, which has been produced by distillation in a vacuum, shows no δ-emission when bombarded by α-rays; but when the surface layers absorb gases, δ-rays make their appearance to a greater and greater extent. The velocities of emission of the individual δ-rays vary extensively. The distribution curve, or the curve which represents the frequency of occurrence as a function of the velocity, is, on the other hand, independent of the velocity of the exciting α-rays. Particles with a velocity corresponding to 2 volts occur with the greatest frequency. Particles of 15 volts velocity account for about 1 per cent. of the total radiation, but velocities as high as 2,400 volts can also be detected.

In consequence of the occurrence of δ-rays, the α-ray tracks produced by the cloud method no longer appear as single lines of fog, but they reveal fine secondary ionization tracks emitted from the main track in all directions (cf. Fig. 27, Plate III).

The occurrence of δ-rays is largely responsible for the fact that saturation is attained only at high potentials for an ionization current (cf. p. 10) due to α-rays. The ion density along the track of the α-particle is especially large, so that we may speak of columnar ionization. In this we have ionization not only of the molecules directly struck by the α-particle, but the δ-rays released in the process also contribute fundamentally to the ionization produced.

If α-particles impinge on atoms of hydrogen, these are ionized and acquire very large velocities as a result of the collision, since their mass is only one-quarter of that of α-particles. The range of these particles is also several times that of the α-particles producing them. In this way 'H-rays' are produced in hydrogen by the α-particles from RaC$'$, the range of these H-rays amounting to about 28 cm. The number of pairs of ions produced by an H-particle per cm. of its path has a value only one-quarter of that produced by an α-particle of the same velocity.

By means of scintillation observations (see p. 20) it has been possible to detect a small number of particles with a considerably greater range, when various substances are bombarded by α-rays. The observation of such H-rays, which originate in the nuclei of the atoms struck, has been of great importance in the elucidation of the nuclear structure of atoms (see p. 94 et seq.).

When α-particles are incident on lead, tin, etc., they are able to liberate characteristic X-rays of the elements mentioned, but the intensity of the X-radiation produced in this way is very feeble. Thus from the K-level of zinc only 0·00023 of a quantum of K-radiation is liberated along its track by an α-particle from polonium, and the yield in the excitation of L-rays is still smaller. Moreover, a γ-radiation has been observed to occur during the bombardment of Li, Be, B, F, Mg, and Al with α-rays. The hardness of these rays is of the same order of magnitude as that of the hardest γ-rays, but their intensity is exceedingly small. (Cf. p. 102.)

REFERENCES

A detailed discussion of the properties of the secondary radiations will be found in the *Handbuch der Physik*, 2nd ed. (Springer, Berlin, 1933), vols. xxii/2 and xxiii/2. X-ray spectroscopy is fully dealt with in *Spektroskopie der Röntgenstrahlen* 2nd ed.), by M. SIEGBAHN (Berlin, 1931); an English translation of the 1st edition was published by the Clarendon Press, Oxford (1925).

VI

THE RECOIL RAYS

When an α-particle is emitted from an atom, the latter recoils in the opposite direction in accordance with the principle of action and reaction, just as a gun recoils when a shot is fired from it.

If the mass and velocity of the projectile are m_1 and v_1, and those of the gun m_2 and v_2, we have

$$v_2 = v_1 . \frac{m_1}{m_2}.$$

Thus an atom of RaA emits α-particles with a velocity of $1{\cdot}69 \times 10^9$ cm./sec., and is transformed at the same time into an atom of RaB, which is projected by virtue of recoil with a velocity of

$$1{\cdot}69 \times 10^9 \times \frac{4}{214} = 3{\cdot}31 \times 10^7 \text{ cm./sec.},$$

the atomic weight of the α-particle being 4, and that of the RaB atom being 214. The velocity of the residual atom of RaB is considerably less than that of the α-particle, but it is sufficient to ionize the air along its track, to act on a photographic plate, etc. As a consequence of the smaller velocity of the recoil rays, the number of ions produced by each of them per cm. of its track is, in fact, markedly greater than and reaches a value five times as large as that due to an α-particle. As with the α-particles, the ability of the recoil particle to produce these effects ceases suddenly, the range in air amounting to about 0·14 mm., and in silver to 2×10^{-5} mm. Owing to their smaller velocity, the phenomenon of scattering occurs in a more marked degree with recoil rays than with α-particles. From the magnitude of the deflexion in a magnetic field it follows that the recoil atom has an atomic weight of about 214, which agrees well with that of RaB, and that it carries a single elementary charge (cf. p. 70).

If we have a deposit of RaA on a horizontal metal plate, the atoms that emit α-particles in an upward direction will be projected downwards, and will penetrate the upper layers of the plate; conversely, those atoms that have emitted α-particles in the downward direction will be projected upwards into the air surrounding the plate. The most favourable yield of recoil atoms collected after leaving the plate by recoil accordingly amounts to 50 per cent.

The possibility of obtaining individual radio-elements in a pure state by utilizing the phenomenon of radioactive recoil plays a very important role in the study of radioactivity (see p. 218). In order to be independent of the small range of the recoil atoms, they are collected in an electric field, i.e. the preparation is raised to a positive potential of say 100 volts, the negative pole of the battery being connected to an insulated plate fixed a few millimetres above the preparation. Except for a very small fraction the residual atoms are positively charged, and hence they are drawn through the air to the upper plate (see Fig. 28). After emission of the positively charged α-particles, the recoil atoms are at first negatively charged, but when they ionize molecules of the surrounding air they part with several electrons and so themselves acquire a positive charge. An investigation of the deflexion of the recoil atoms of RaB in a magnetic field has revealed that practically all of them carry a single positive charge, and that only about 0·001 per cent. of them are negatively charged.

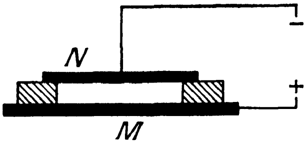

Fig. 28. Arrangement for collecting Recoil Atoms.

The phenomenon of recoil must also occur during the emission of a β-particle, but the resulting residual atoms are much more difficult to detect than the products of α-recoil. The kinetic energy of the projected particle is here much smaller, and is usually insufficient to propel the particle away from the surface of the plate on which the disintegration takes place. It corresponds to the kinetic energy of an equally heavy singly charged ion that has traversed a potential difference of a few tenths of a volt. Nevertheless, it is possible to collect up to 23 per cent. of the RaC atoms which suffer recoil in the disintegration of RaB, by the application of special precautionary measures. These include working in a high vacuum, purification of the initial substance by distillation in a vacuum, deposition of the material used on a polished surface that has been cooled to a low temperature to prevent the volatilization of the particles.

The photographic detection of recoil atoms is only possible when the gelatine layer of the plates is so thin that the recoil atoms are not brought to rest before they reach the silver bromide.

We meet with a special type of recoil in the phenomenon of 'aggregate recoil'. If we deposit polonium electrolytically on a bright metal foil and place a second foil near to and facing it, we can detect polonium also on the second foil after a short time. The explanation of this phenomenon is that, during electrolysis, the polonium is not deposited on the foil in the form of individual atoms, but as aggregates of two or more atoms. When one of the atoms of such an aggregate disintegrates, not only the RaG atom resulting from the disintegrated polonium suffers recoil, but the unchanged polonium atoms present in the same aggregate are projected with it from the foil. Thus when an α-particle is emitted in the direction of the first foil, the remaining aggregate, consisting of a RaG atom and one or more polonium atoms, is projected in the direction of the second plate. This phenomenon supplies an explanation of the fact that all objects in the neighbourhood of strong polonium preparations become contaminated with polonium, even though this element gives off no emanation, and its product of disintegration is no longer active. Aggregate recoil phenomena can also be observed with preparations in which the polonium was not deposited electrolytically, but by collecting the active deposit from radium emanation, and other radio-elements that emit α-rays show similar effects. Aggregates of the active deposit are also produced in mixtures of emanation and air or argon.

REFERENCES

The properties of recoil particles are fully discussed in *Radiations from Radioactive Substances*, by Rutherford, Chadwick, and Ellis (Cambridge University Press, 1930), and in *Radioaktivität*, by Meyer and Schweidler (Teubner, Leipzig, 1927).

VII

POSITRONS AND NEUTRONS

A. Positrons

Positive electrons are not emitted in natural radioactive processes, but the emission of such particles can be detected in the disintegration of different products of artificial radioactivity (see p. 120). Thus aluminium (cf. p. 120, Table 30) that has been rendered active artificially by bombardment with α-rays emits positive electrons or 'positrons', and gradually loses its activity in the process. The positrons emitted by aluminium have very different velocities; they show a continuous energy spectrum with a limiting maximum energy of about 3×10^6 e.V., the most frequently represented energy value being 9×10^5 e.V. The mass, charge, and ionizing power of the positron have been found to be the same as those of the electron. Apart from the sign of the charge, however, there is found to be a fundamental difference between the electron and the positron, for positrons have no permanent existence. When they collide with electrons they combine with them, and the energy of 10^6 e.V. liberated in the process of union is emitted in the form of quanta of γ-rays. In most cases the union is effected with electrons at rest, and then two quanta each of energy 500,000 e.V. are emitted in opposite directions; but there are also processes in which the total energy of annihilation is emitted in a single quantum of energy 10^6 e.V. Accordingly, the incidence of positrons on matter results in the emission of a γ-radiation (cf. p. 52) which is independent of the nature of the absorbing element; in all cases we are concerned solely with the combination of positrons with electrons of the element under consideration. Conversely, positrons may be produced by the incidence of hard γ-rays on matter, e.g. the γ-rays from RaC, the energy of which lies between $1{\cdot}0\times10^6$ and $2{\cdot}2\times10^6$ e.V. Positrons are also released by the action of electrons when their energy exceeds 10^6 e.V.

Positrons were first observed in the examination of the cloud tracks produced in a Wilson chamber by the cosmic rays, when individual tracks were found to have been deflected by the magnetic field in a direction opposite to those due to electrons.

It was found recently that RaC and (ThC+ThC″) emit a very small number of positrons, to wit, one positron to every 3,000 disintegrating atoms. These positrons are not emitted by the nucleus of the disintegrating atom, but are the result of secondary processes [1].

B. NEUTRONS

As with positrons, neutrons do not appear in the course of natural radioactive disintegration processes; on the other hand, they are frequently observed in processes of constrained transformation of the elements. Thus when α-particles strike beryllium nuclei, beryllium is transformed into carbon, and at the same time neutrons are liberated in accordance with the equation:

$$^{9}_{4}\mathrm{Be}+^{4}_{2}\mathrm{He} = ^{12}_{6}\mathrm{C}+^{1}_{0}n.$$

This process is described in greater detail on p. 78.

Neutrons are atomic nuclei which possess no charge, as it were atoms of the element of zero atomic number. Since they possess no charge, they do not interact with the peripheral parts of atoms with which they collide, and hence they do not effect ionization in their passage say through air; because of this they cannot be detected by methods similar to those used for the other types of rays, already described. However, neutrons do interact with atomic nuclei, and by virtue of this interaction they can be detected. When neutrons collide with hydrogen nuclei the latter are set into rapid motion, and these moving protons can be detected, e.g. by use of a cloud chamber. The detection of fast neutrons is effected by making use of an electroscope filled with hydrogen or lined with paraffin wax, and also by their ability to bring about artificial radioactivity (cf. p. 114). If we are faced with the problem of deciding whether or not fast neutrons are being emitted by a source, it suffices to introduce say silicon in the form of a glass plate into the path of the supposed neutrons, and then after some time to test the glass plate by bringing it near to a tube counter. Neutrons disrupt the silicon nucleus and call forth an artificial radioactivity, which decays under emission of hard β-rays (cf. p. 116). From the occurrence of these readily detectable β-rays we can infer the presence of fast neutrons. If we require to detect slow neutrons, iodine, silver, rhodium, or dysprosium may be introduced into the path of the supposed neutrons. These elements serve as specially suitable 'receivers' for slow neutrons, and the capture of the neutrons brings about in them an easily detectable artificial radioactivity. Another method that is likewise much used consists in allowing the neutrons to enter an ionization chamber, the walls of which are coated with boron or lithium. The following processes take place under the action of slow neutrons:

$$^{10}_{5}\mathrm{B}+^{1}_{0}n = ^{7}_{3}\mathrm{Li}+^{4}_{2}\mathrm{He} \quad \text{and} \quad ^{6}_{3}\mathrm{Li}+^{1}_{0}n = ^{3}_{1}\mathrm{H}+^{4}_{2}\mathrm{He}.$$

The energetic particles produced in this way ionize the gas contained in the ionization chamber. If the capture of a neutron leads to the formation of a stable isotope and not to a radioactive isotope of the element concerned, it is then impossible to resort to artificial radioactivity for the detection of the neutrons. An example of this is the capture of a neutron by lead 206, whereby the stable isotope of lead (207) is formed in accordance with the equation:

$$^{206}_{82}\text{Pb}+^{1}_{0}n = ^{207}_{82}\text{Pb}.$$

In such cases the capture of neutrons can be detected by the emission of γ-rays. The incorporation of neutrons into atomic nuclei is always an exothermic process, and the energy liberated is emitted in the form of γ-rays.

1. Absorption and Scattering of the Flux of Neutrons

The flux of neutrons is reduced owing to their gradual capture by the nuclei of the atoms penetrated; a reduction also arises from elastic collisions of the neutrons with atomic nuclei, and by nuclear scattering. Neutrons suffer elastic collisions in particular with protons, e.g. in water or in paraffin wax, and this leads to a rapid decrease in the energy of the neutrons. When the velocity of a neutron is markedly greater than the mean velocity of the molecules of a gas, and it collides with a proton, the energy of the neutron is reduced on the average to one-half of its initial value. Accordingly, after ten collisions with protons the mean energy of the neutron will have fallen to one-thousandth of its original value. Neutrons of energy 4×10^{6} e.V. are strongly represented in the flux of neutrons emitted by mixtures of radon and beryllium powder, this being the most familiar source of neutrons. After ten collisions such neutrons have on the average an energy of only about 4,000 e.V., and less than thirty collisions with protons are necessary in order to obtain neutrons with velocities of normal molecular magnitude. The mean life of such a neutron in paraffin wax is 2×10^{-4} sec. [3]. One of the ways of demonstrating that the slowest neutrons actually have velocities comparable with those of gaseous molecules is as follows. It has been possible to show that, when paraffin wax is reduced to the temperature of liquid air, slow neutrons suffer a greater reduction of velocity in it, and they are then able to activate silver more strongly than before, since the capture of neutrons by silver and similar nuclei (cf. p. 77) takes place more readily the slower the neutrons. The large diminution in energy experienced by neutrons when they collide with protons is to be attributed to the fact that the mass of a neutron (cf. p. 77) is practically equal to that of a proton, and in consequence of this the transmission of energy from the colliding particle to that struck takes place very readily. The velocity of neutrons can also be reduced by their collision with the nuclei of other light elements such as carbon or silicon, but with them the effect is very much smaller. The velocity of fast neutrons is also reduced by their inelastic collision with heavy atoms, but in this case the energy given off by the neutrons is utilized to excite the heavy nucleus, and it is later emitted in the form of γ-radiation.

In what follows we shall consider separately the reduction in the flux of fast and of slow neutrons, i.e. their absorption and scattering. We know the cross-sections of light nuclei (cf. p. 79), and we can accordingly calculate the reduction in flux which fast neutrons should experience in their passage through light atoms, on the assumption that the whole nuclear cross-section is effective in the scattering.† The values determined experimentally agree approximately with those calculated. When we pass over to heavier nuclei the cross-sections increase progressively. Here it is not possible to compare the cross-sections obtained from the scattering of neutrons with values obtained by other means, since methods which permit the calculation of the cross-sections of light atoms fail in the case of heavy atoms. It is probable that the nuclear cross-section resulting from the 'absorption' values of fast neutrons in their passage through the atoms concerned represents the true nuclear cross-section. The effective cross-sections for fast neutrons lie between $1{\cdot}6 \times 10^{-24}$ cm.^{2} (light nuclei) and $5{\cdot}8 \times 10^{-24}$ cm.^{2} (mercury) [1], and show a fairly regular increase within that range.‡

Slow neutrons show quite a different behaviour from fast ones. The effective cross-section in the scattering of slow neutrons is roughly equal to the total nuclear cross-section. Hydrogen nuclei alone behave differently, for they scatter very much more strongly than would be expected from the magnitude of the cross-section of hydrogen nuclei. The reduction in intensity arising from the absorption and scattering experienced by slow neutrons in nuclei is very different from element to element, and the effective cross-section varies between $1{\cdot}5 \times 10^{-24}$ cm.^{2} for aluminium and $25{,}000 \times 10^{-24}$ cm.^{2} for gadolinium. Cadmium, europium, samarium, and especially gadolinium have large effective cross-sections in respect of the absorption of slow neutrons, as may be seen from Table 20. The effective cross-sections are dependent on the velocity of the neutrons. In the cases of lithium and boron the conditions are particularly simple, for here the effective cross-sections are inversely proportional to the velocity of the neutrons [2]. The large effective cross-section of the cadmium nucleus, on the other hand, is only maintained for neutrons whose energy does not exceed 1 e.V., i.e. for those neutrons of velocity at most 3 to 4 times greater than the molecular velocities met with at room temperatures, in accordance

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† The effective cross-section is equal to the mass absorption coefficient divided by 6×10^{23}, and multiplied by the atomic weight (cf. p. 51).

‡ In all cases the cross-section is due mainly to scattering, for only a small fraction of fast neutrons is captured in the collision process.

with the kinetic theory of gases. For this reason cadmium is used as a screening material whenever it is desired to exclude neutrons with thermal energies. Neutrons with molecular velocities, though powerfully absorbed by cadmium, are only feebly absorbed for example by arsenic. For a particular range of velocity, however, this latter element manifests a large effective cross-section of about 100 times the nuclear cross-section, and within this range it is effective to a degree comparable with that of cadmium for neutrons with molecular velocities. Gold, silver, rhodium, iodine, indium, and possibly many other elements likewise possess a very high absorbing power for a limited range of velocity of the neutrons. This kind of absorption is generally known as 'resonance capture'.

TABLE 20

Nuclear Cross-sections with respect to Slow Neutrons ($cm.^2 \times 10^{-24}$)

Element	*Cross-section*	*Element*	*Cross-section*	*Element*	*Cross-section*
H	48	Co	35	Ba	140
B	500	Ni	15·4	Sm	4,000
C	3·4	Cu	7·4	Eu	3,700
N	11·3	Zn	4·7	Gd	25,000
O	3·3	Br	11·8	Dy	1,300
Na	4·2	Y	119	Ir	285
Mg	3·0	Mo	7·1	Pt	25
Al	1·0	Rh	115	Au	88
Si	2·5	Pd	10	Hg	380
Cl	39	Ag	55	Pb	8·6
K	8·2	Cd	3,300	Th	32
Ca	11	I	9·4	U	43

The production of slow neutrons is effected most simply by surrounding the source of neutrons with paraffin wax [3], the source being say a glass tube containing a mixture of radon and beryllium. By frequent collision of the fast neutrons with paraffin wax their velocity is reduced to molecular magnitude, and these retarded neutrons further diffuse until they either emerge from the paraffin block or are absorbed by a hydrogen nucleus. Neutrons with molecular velocities can only diffuse through a distance of about 2 to 3 cm. in paraffin wax, and for this reason slow neutrons with the velocity of gaseous molecules emerge only from the outer layer of thickness 2 to 3 cm. of a large cylinder of paraffin wax. When large cylinders of paraffin wax are used, the yield of slow neutrons is small, and diminishes further as the circular dimensions are increased. Likewise, if the cylinder is chosen so that its radius is less than 2 to 3 cm., a poor yield of slow neutrons is obtained, since the quantity of paraffin wax does not then suffice to retard the fast neutrons adequately. The alteration in the number of slow neutrons that strike a surface of silver in unit time when the thickness of the cylindrical wax layer surrounding the source is varied follows from a consideration of Fig. 29 [4]. The slow neutrons are captured by the silver nuclei, and bring about artificial radioactivity in the silver. The activity is proportional to the number of slow neutrons incident on the silver, and in the graph the activity of the silver is plotted in its dependence on the thickness of the wax layer surrounding the source of neutrons. The maximum yield is obtained when the thickness of the layer of paraffin wax is about 9 cm. If we wish to produce neutrons of intermediate velocity, we must choose a thinner layer of paraffin wax. Elements that show no marked absorbing power for neutrons scatter them to a considerable degree. In experiments with neutrons, even the scattering effected by the walls of the room may have a disturbing influence.

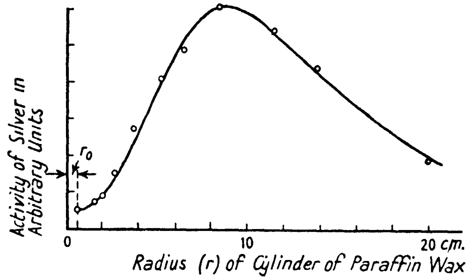

FIG. 29. Activity induced in silver situated at a fixed distance from the neutron source, surrounded by wax cylinders of different radius (r).

2. The Mass of the Neutron

Since a neutron is not deflected in an electric or in a magnetic field, the methods which served for the determination of the masses of α- and β-particles are of no avail here. The following method is the simplest and most accurate for the evaluation of the mass of the neutron [5]. Under the action of γ-rays, neutrons (n) are split off from the atoms of heavy hydrogen, deuterium (D), in accordance with the equation:

$$ {}^{2}_{1}\mathrm{D}+E_{\gamma} = {}^{1}_{1}\mathrm{H}+{}^{1}_{0}n+E_{\mathrm{H}}+E_{n}. $$

Since the energy E_{γ} of the γ-ray quanta of ThC″ from radiothorium used in the experiments amounts to $2{\cdot}65\times10^{6}$ e.V., corresponding to 0·00285 unit of atomic weight,† and the kinetic energies E_{H} and E_{n} of the respective protons and neutrons produced both amount to $0{\cdot}25\times10^{6}$ e.V., the above equation takes the following form:

$$ 2{\cdot}01471+0{\cdot}00285 = 1{\cdot}00812+0{\cdot}000269+0{\cdot}000269+n, $$

$$ \text{or}\quad n = 1{\cdot}00890. $$

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† 1 electron-volt = $1{\cdot}0741\times10^{-9}$ unit of atomic weight.

The smallest amount of energy necessary to split off a neutron from the deuterium atom is $2{\cdot}1\times10^6$ e.V., and for the atom ^{9}Be $1{\cdot}6\times10^6$ e.V. From amongst all atomic nuclei, that of ^{9}Be is found to have the most weakly bound neutrons. In the production of neutrons by bombardment with α-rays, beryllium is for this reason to be preferred to all other elements, as also when the neutrons are released by the aid of the γ-rays from radium. On the other hand, when radiothorium is used as a source of γ-rays, six times as many neutrons are emitted by deuterium as by beryllium. The greatest yield of neutrons is attained in the bombardment of compounds containing heavy hydrogen (e.g. ND_4Cl) by means of positive rays of deuterium.

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VIII

CONSTITUTION OF THE ATOM AND RADIOACTIVE RAYS

The behaviour of the rays from radioactive substances in their passage through matter has been our most reliable guide in the investigation of the constitution of atoms; conversely, the theory of atomic structure has cleared up many points on the origin and nature of the individual types of rays. Accordingly, we shall now give a short account of the constitution of the atom.

1. The Constitution of the Atom

It follows from the behaviour of α- and β-particles in their passage through atoms that the material part of the atom does not embrace the whole of the atom, regarded as a sphere of radius 10^{-8} cm., but is restricted to an exceedingly small space at the centre of the atom, of diameter about 10^{-13} cm.† Moreover, this atomic nucleus is positively charged. The remainder, or practically the whole of the space inside the atomic sphere is at the disposal of the electrons that revolve round the nucleus. The dimensions of this electron cloud determine the magnitude of the atom, and the number of electrons in it is equal to the number of elementary units of positive charge associated with the nucleus. As a whole, the atom thus appears electrically neutral.

The total number of electrons revolving in the atom, and hence also the number of nuclear positive charges, is equal to the atomic number (see p. 156) of the element concerned. The number of orbital electrons is thus 1 for the hydrogen atom, 2 for helium, 3 for lithium, 29 for copper, 78 for platinum, and 92 for uranium, the element of highest atomic number.

If the electrically neutral atom becomes ionized, it assumes either a positive or a negative charge. In the former case the outer electron layer loses a definite number of electrons, and in the second case the atom takes up one or more electrons from the surrounding matter or from the free electrons in the surrounding space. Ionization or chemical change of any kind, however, can produce no alteration in the number of charges on the nucleus, and hence these processes are always reversible. The reversibility of all changes in the character of the atom produced by chemical processes signifies nothing else than the long-known law

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† This value is derived from the magnitude of the deviation suffered by α-particles when they penetrate the vicinity of the nucleus (see p. 95). The radius of the sun is about 6,500 times smaller than that of the orbit of Neptune; the nucleus of the atom, which resembles in its structure a solar system, is about 10,000 times smaller than the distance of the electrons from the centre of the system.

of the conservation of the elements. In the relatively rare cases of radioactive processes on the other hand, the nucleus, and hence also the system of electrons surrounding it, undergo permanent transformation. The emission of the rays from radioactive substances is very intimately related to this transformation, which, as described on p. 154, involves a complete alteration in the chemical nature of the atom.

The stability of an atom built up in the manner indicated cannot be explained on the basis of classical mechanics and electrodynamics. In order to understand it, it was necessary to formulate the hypothesis that there are certain privileged states of motion of the atom, in which states the atom persists without loss of energy. The energy content of the atom can change only when the atom passes over from one such privileged state into another (see p. 90).

For the interpretation of radiation processes it was necessary to supplement this hypothesis by a second one, according to which, if such a transition is associated with the emission of radiation, the latter is always monochromatic, and of frequency equal to the quotient of the amount of energy radiated and Planck's constant h.

The experimental evidence in support of the view outlined above is so multifarious that it cannot be treated exhaustively here. We need only mention the following especially important phenomena: the scattering of the α-rays in their passage through matter (p. 31), isotopy (p. 147), and the emission and absorption of light, X-rays, etc., as they are revealed in the spectra of the elements.

Before we proceed to the question of the origin of the rays from radioactive substances, it will be advisable to discuss in somewhat greater detail the structure of the atom in the simplest cases, by first making use of the original presentation due to Bohr.

2. Structure of the Hydrogen Atom

A hydrogen atom consists of a nucleus, carrying only a single positive charge, and an electron, which, in accordance with the first hypothesis mentioned above, always moves in certain permissible orbits. This motion is subject to Kepler's laws, like that of a planet round the sun, which is situated at the focus of the orbit, so that the privileged orbits of the electron are ellipses.

There is quite a number of such stationary orbits, but one of them is unique in that when the electron is moving in it, the atom possesses its smallest content of energy. The hydrogen atom can persist permanently in this 'normal state', whereas the 'life' of all other stationary states is very short.

According to the second hypothesis mentioned above, a monochromatic pulse of waves is emitted when an electron passes over from one stationary state to another. In Fig. 30 we see the spectroscopic consequences of the return of an electron to its normal orbit from another stationary orbit, into which it has passed by virtue of some outside influence such as bombardment by cathode rays. When the return to the normal orbit takes place from orbit 2, a spectral line of wave-length 121 mμ is emitted, whilst if the transition takes place from orbit 3, a line of shorter wave-length 102 mμ is produced, because of the greater energy difference between the two orbits, and so on for the other transitions. Should the return take place from an infinitely distant orbit, a spectral line of wave-length 91 mμ is emitted. This last case is equivalent to the 'neutralization' of a hydrogen ion, since of course the converse process of the removal of an electron to infinity constitutes the phenomenon of 'ionization'. In the light of the theory of series spectra, this neutralization process thus appears as a limiting case of the discontinuous quantum states. The above-mentioned lines (Fig. 30) all lie in the extreme ultra-violet. The lines of the visible spectrum of hydrogen likewise have their origin in transitions from one transitory orbit to another, e.g. from orbits 3, 4, etc. to orbit 2.

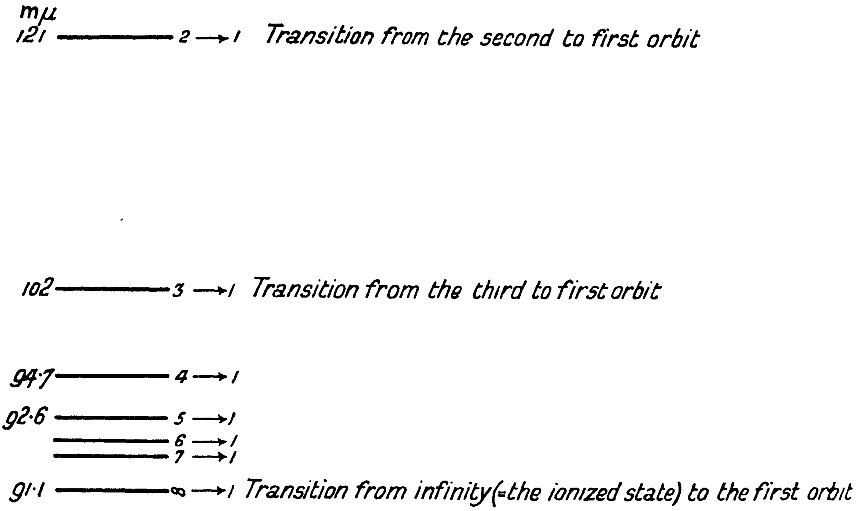

FIG. 30. Spectrum of Hydrogen in the Ultra-violet Region (Lyman Series).

The relative distances of the individual stationary orbits from the centre of the atom are represented in Fig. 31. The diameter of the first orbit is that of the atom in its normal state, and is equal to $1{\cdot}06 \times 10^{-8}$ cm. The calculation of the orbital radii involves only three universal constants: the elementary electrical charge e, the mass of the electron m, and Planck's constant h (p. 62). The same quantities also suffice to calculate the work that is necessary to transfer an electron from one stationary orbit to another. For instance, we can calculate the work necessary to remove entirely an electron from the atom, and when the result is expressed in volts (see p. 43), we find the value to be 13·5; this is the ionization potential of the hydrogen atom.

In the construction of Fig. 31 it was assumed, for purposes of simplicity, that the electron moves in circular orbits, so that in order to differentiate between the stationary orbits or to describe the processes one number suffices, this being called the quantum number. In order to be able to describe the spectral behaviour in all its details, however, it was necessary to introduce a second, and later still a third and even a fourth quantum number. Even when a motion takes place in stationary elliptical orbits, it is no longer sufficient to characterize the stationary orbit by one number; recourse must be had to a second integral number to differentiate between the different eccentricities of the orbits. It will be clear from Fig. 32 that the major axes of the ellipses have the same length for orbits which possess the same first quantum number. The minor axes vary with the second quantum number in such a way that the orbit has its greatest eccentricity when the second quantum number is equal to unity, whilst the orbit is circular when the first and second quantum numbers have the same value.

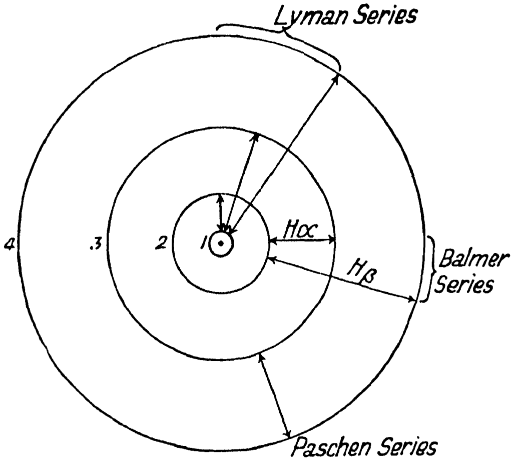

Fig. 31. Stationary Circular Orbits in the Hydrogen Atom.

The same theory which led to our knowledge of the structure of the hydrogen atom has been applied also to obtain information on the structure of the other atoms. At the same time, this work was greatly aided by the results of spectroscopic research, of X-ray spectroscopy, by investigations on the bombardment of atoms by electrons, and by research in chemistry. It has been found that also when several electrons are present in the atom, the orbit of every electron is characterized by quantum numbers, in a manner analogous to the possible orbits in the hydrogen atom.

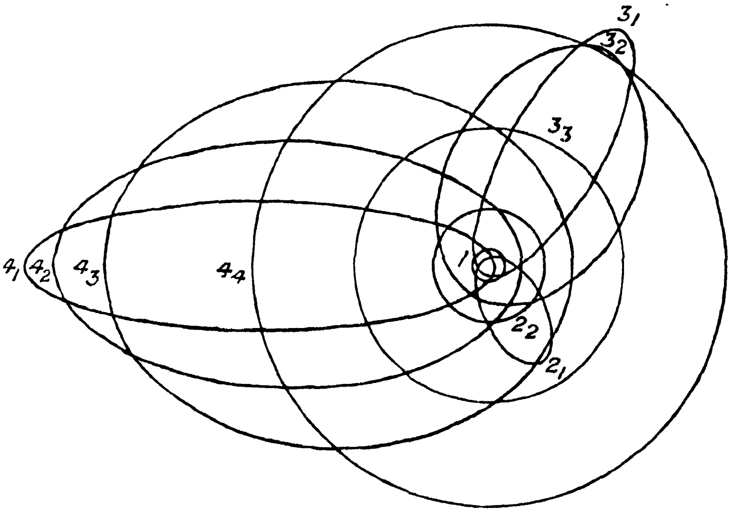

FIG. 32. Stationary Elliptical Orbits in the Hydrogen Atom.

3. Atoms of Higher Atomic Number

The following symbols are used for the principal quantum number n:

K	*L*	*M*	*N*	*O*	*P*
1	2	3	4	5	6;

whilst the azimuthal quantum number l is characterized by the symbols:

s	*p*	*d*	*f*
0	1	2	3.

Many lines which appear as single lines in spectroscopes of average resolving power are found to be double lines or groups of neighbouring lines when examined with apparatus of high resolving power. In the accurate description of this multiplicity of spectral lines a further (third) quantum number is introduced, the physical significance of which is as follows: The electrons exhibit a rotation proper to themselves and generally referred to as 'spin', and this confers upon them the property of small magnets. In the case of a single electron the strength of binding is somewhat different, according to the direction of the spin. Two opposed possibilities present themselves for the spin, so that for an electron with the same principal and azimuthal quantum numbers there are two different values of the energy of binding, whence the doublets in the spectra of the alkalis. If instead of one electron we are dealing with two mutually interacting electrons, the conditions are more complicated, in the first place by the spin, in the manner indicated, and furthermore by the different possibilities of orientation of the electron orbits with respect to each other. In this way arise the triplets of the alkaline earths and the still higher multiplets of the more complicated atoms.

In strong magnetic fields spectral lines are resolved into components, the theoretical interpretation of which requires the introduction of a fourth 'magnetic quantum number' m. In a strong magnetic field the resultant vectors of electron spin and orbital impulse experience a definite directional quantization. In order to establish the distribution numbers of the level with a given principal quantum number n it suffices, in the first place, to disregard spin. In this case the number of possible orientations is one for an s-electron ($l = 0$), three for a p-electron ($l = 1$), five for a d-electron ($l = 2$), and in general $(2l+1)$ for an azimuthal quantum number l. The magnetic quantum number can accordingly assume $(2l+1)$ different values and we must assign to it all the whole numbers from $-l$ to $+l$. Table 21 shows the possible combinations of the three quantum numbers n, l, m, without taking into consideration the spin quantum number. Each of these combinations may occur in two ways, either with the spin quantum number $+\frac{1}{2}$ or $-\frac{1}{2}$. Since the spin quantum numbers have to differ by unity and be equal in amount, they can only assume the values $+\frac{1}{2}$ and $-\frac{1}{2}$. In the unexcited hydrogen atom, in the ground state where $l = s = 0$, the first horizontal row of Table 21 reveals to us only one possible combination, but this is possible in two ways, with the spin quantum number $+\frac{1}{2}$ or $-\frac{1}{2}$. There are accordingly two kinds of electrons present in normal

TABLE 21

Combinations of the Principal, Azimuthal, and Magnetic Quantum Numbers

$n = 1$	$l = 0$	$m = 0$
$n = 2$	$l = 0$	$m = 0$
$n = 2$	$l = 1$	$m = -1\ 0\ +1$
$n = 3$	$l = 0$	$m = 0$
$n = 3$	$l = 1$	$m = -1\ 0\ +1$
$n = 3$	$l = 2$	$m = -2\ -1\ 0\ +1\ +2$
$n = 4$	$l = 0$	$m = 0$
$n = 4$	$l = 1$	$m = -1\ 0\ +1$
$n = 4$	$l = 2$	$m = -2\ -1\ 0\ +1\ +2$
$n = 4$	$l = 3$	$m = -3\ -2\ -1\ 0\ +1\ +2\ +3$

hydrogen atoms. For a p-state of the atom ($l = 1$) there are already six possible combinations, and so on. Table 22 shows the grouping of electrons in the whole periodic system.

The final fixing of the numbers was facilitated by the application of Pauli's principle. According to this, two electrons in an atom can never agree in all four quantum numbers, and hence the combinations given in the individual horizontal rows of Table 21 can be realized by at most two electrons, to wit, one with a right-handed and one with a left-handed spin. The application of Pauli's principle makes it clear

TABLE 22

The Grouping of Electrons in the Periodic System

	K	*L*		*M*			*N*				*O*			*P*			*Q*
	s	*s*	*p*	*s*	*p*	*d*	*s*	*p*	*d*	*f*	*s*	*p*	*d*	*s*	*p*	*d*	*s*
1 H	1																
2 He	2																
3 Li	2	1															
4 Be	2	2															
5 B	2	2	1														
6 C	2	2	2														
7 N	2	2	3														
8 O	2	2	4														
9 F	2	2	5														
10 Ne	2	2	6														
11 Na	2	2	6	1													
12 Mg	2	2	6	2													
13 Al	2	2	6	2	1												
14 Si	2	2	6	2	2												
15 P	2	2	6	2	3												
16 S	2	2	6	2	4												
17 Cl	2	2	6	2	5												
18 A	2	2	6	2	6												
19 K	2	2	6	2	6		1										
20 Ca	2	2	6	2	6		2										
21 Sc	2	2	6	2	6	1	2										
22 Ti	2	2	6	2	6	2	2										
23 V	2	2	6	2	6	3	2										
24 Cr	2	2	6	2	6	5	1										
25 Mn	2	2	6	2	6	5	2										
26 Fe	2	2	6	2	6	6	2										
27 Co	2	2	6	2	6	7	2										
28 Ni	2	2	6	2	6	8	2										
29 Cu	2	2	6	2	6	10	1										
30 Zn	2	2	6	2	6	10	2										
31 Ga	2	2	6	2	6	10	2	1									
32 Ge	2	2	6	2	6	10	2	2									
33 As	2	2	6	2	6	10	2	3									
34 Se	2	2	6	2	6	10	2	4									
35 Br	2	2	6	2	6	10	2	5									
36 Kr	2	2	6	2	6	10	2	6									

TABLE 22 (*cont.*)

	K	*L*		*M*			*N*				*O*			*P*			*Q*
	s	*s*	*p*	*s*	*p*	*d*	*s*	*p*	*d*	*f*	*s*	*p*	*d*	*s*	*p*	*d*	*s*
37 Rb	2	2	6	2	6	10	2	6			1						
38 Sr	2	2	6	2	6	10	2	6			2						
39 Y	2	2	6	2	6	10	2	6	1		2						
40 Zr	2	2	6	2	6	10	2	6	2		2						
41 Nb	2	2	6	2	6	10	2	6	4		1						
42 Mo	2	2	6	2	6	10	2	6	5		1						
43 ..	2	2	6	2	6	10	2	6	(5)		(2)						
44 Ru	2	2	6	2	6	10	2	6	7		1						
45 Rh	2	2	6	2	6	10	2	6	8		1						
46 Pd	2	2	6	2	6	10	2	6	10								
47 Ag	2	2	6	2	6	10	2	6	10		1						
48 Cd	2	2	6	2	6	10	2	6	10		2						
49 In	2	2	6	2	6	10	2	6	10		2	1					
50 Sn	2	2	6	2	6	10	2	6	10		2	2					
51 Sb	2	2	6	2	6	10	2	6	10		2	3					
52 Te	2	2	6	2	6	10	2	6	10		2	4					
53 I	2	2	6	2	6	10	2	6	10		2	5					
54 X	2	2	6	2	6	10	2	6	10		2	6					
55 Cs	2	2	6	2	6	10	2	6	10		2	6		1			
56 Ba	2	2	6	2	6	10	2	6	10		2	6		2			
57 La	2	2	6	2	6	10	2	6	10		2	6	1	2			
58 Ce	2	2	6	2	6	10	2	6	10	(1)	2	6	(1)	(2)			
..	..	..			..				..			..			..		..
..	..	..			..				..			..			..		..
72 Hf	2	2	6	2	6	10	2	6	10	14	2	6	(2)	(2)			
73 Ta	2	2	6	2	6	10	2	6	10	14	2	6	(3)	(2)			
74 W	2	2	6	2	6	10	2	6	10	14	2	6	4	2			
75 Re	2	2	6	2	6	10	2	6	10	14	2	6	(5)	(2)			
76 Os	2	2	6	2	6	10	2	6	10	14	2	6	(6)	(2)			
77 Ir	2	2	6	2	6	10	2	6	10	14	2	6	(7)	(2)			
78 Pt	2	2	6	2	6	10	2	6	10	14	2	6	8	2			
79 Au	2	2	6	2	6	10	2	6	10	14	2	6	10	1			
80 Hg	2	2	6	2	6	10	2	6	10	14	2	6	10	2			
81 Tl	2	2	6	2	6	10	2	6	10	14	2	6	10	2	1		
82 Pb	2	2	6	2	6	10	2	6	10	14	2	6	10	2	2		
83 Bi	2	2	6	2	6	10	2	6	10	14	2	6	10	2	3		
84 Po	2	2	6	2	6	10	2	6	10	14	2	6	10	2	4		
85 ..	2	2	6	2	6	10	2	6	10	14	2	6	10	2	5		
86 Rn	2	2	6	2	6	10	2	6	10	14	2	6	10	2	6		
87 ..	2	2	6	2	6	10	2	6	10	14	2	6	10	2	6		1
88 Ra	2	2	6	2	6	10	2	6	10	14	2	6	10	2	6		2
89 Ac	2	2	6	2	6	10	2	6	10	14	2	6	10	2	6	(1)	(2)
90 Th	2	2	6	2	6	10	2	6	10	14	2	6	10	2	6	(2)	(2)
91 Pa	2	2	6	2	6	10	2	6	10	14	2	6	10	2	6	(3)	(2)
92 U	2	2	6	2	6	10	2	6	10	14	2	6	10	2	6	(5)	(1)

why the *K*-group can contain only 2 electrons, the *L*-group only 8, the *M*-group only 18, and the *N*-group only 32, and why the periods in the natural classification of the elements contain respectively 2, 8, 18, and 32 elements. For if, in the *K*-, *L*-, *M*-, *N*-, etc., groups, we differentiate sub-groups (s, p, d, f) according to their azimuthal quantum numbers, we obtain as the highest numbers that a sub-group can exhibit (as can be seen from Table 21, p. 84, in which $l = 0 = s$, $l = 1 = p$, $l = 2 = d$, $l = 3 = f$):

2 electrons for an s-sub-group (taking account of spin),
6 electrons for a p-sub-group,
10 electrons for a d-sub-group,
14 electrons for an f-sub-group.

Now in the K-group the azimuthal quantum number is always zero, i.e. only one s-sub-group is present, in the L-group only one s- and one p-sub-group, and so on. We thus obtain as the maximum numbers of the individual groups:

TABLE 23

Maximum Number of Electrons in the Sub-groups

2 in the one-quantum K-group
$2+6 =$ 8 in the two-quantum L-group
$2+6+10 =$ 18 in the three-quantum M-group
$2+6+10+14 =$ 32 in the four-quantum N-group

The maximum numbers 2, 8, 18, 32 are the same as characterize the lengths of the individual periods in the natural classification of the elements.

In the light of the views outlined above we shall now briefly compare the origin of the characteristic X-ray spectrum with that of the optical series spectra. Whereas, as discussed above, the optical spectrum of an element owes its origin to an electron from the outer shell of the atom jumping from one possible position to another, the sudden change in position of electrons in the inner layers nearest the nucleus gives rise to the production of the X-ray spectrum. Compared with that on the outer electrons, the action of the nucleus on the inner electrons is so overwhelming that it can be considered as almost solely operative. It is due to this circumstance that simple relationships are found between the characteristic X-ray spectra of the elements and their atomic numbers.

On the other hand, the processes which take place in the outer layers, well removed from the nucleus of the atom, are scarcely subject to a direct influence of the nucleus, and for this reason we find here no simple relation between the nuclear charge and the optical spectrum. The position of an element in its group is the determining factor for its optical spectrum; thus the spectra of the alkali metals show an essentially similar type of structure, in spite of their markedly different atomic numbers, because the peripheral arrangement of their electrons is the same.

One of the most striking differences between X-rays and optical rays is to be found in the fact that no absorption lines occur in the X-ray spectrum. Thus when white light passes through sodium vapour, the *D*-lines, which are characteristic emission lines of sodium, are absorbed by the vapour, and reveal themselves as sharp absorption lines in the spectrum. The analogous phenomenon does not occur in the case of X-ray lines; the K_α-line of an element undergoes no greater absorption by a layer of the same element than the X-ray lines of only slightly greater or slightly lesser frequency. The absorption rises suddenly only when appreciably shorter wave-lengths are used, and then the K_α-line is simultaneously emitted. This characteristic X-radiation of the elements is also sometimes called 'fluorescent' radiation, because in optics we find a similar behaviour with fluorescent substances, which likewise convert light of shorter wave-length into light of longer wave-length.

On the basis of our conceptions of atomic structure this difference becomes clear. Before an X-ray absorption line could arise, an electron from one of the innermost orbits of the atom would have to be removed into a more distant orbit; but all the orbits near the nucleus have their full complement of electrons, and absorption can only take place when the energy corresponding to the incident wave-length suffices to remove an inner electron to the periphery of the atom. To supply the necessary energy for this purpose, a distinctly shorter wave-length is required than corresponds to the emission lines of the element, in contrast to the case of optical absorption spectra, for the production of which it is only necessary to raise outer circulating electrons to a slightly higher energy level. But once an electron has been removed from the innermost orbit to beyond the surface of the atom, in the manner described, the resulting gap will be filled by an electron from the second, or third, or higher orbit, but not from the surface of the atom, and in this way arise respectively the emission lines K_α, K_β, and so forth.

The mode of production of the X-ray spectrum is very clearly revealed in the diagram of Fig. 33. The *K*-excitation consists in the removal of an electron from the *K*-group; in the diagram this is indicated by the arrow between *K* and the normal state. The resulting gap in the atom, due to the incomplete population of the *K*-shell, may now be filled by the transition of an electron from the *L*-level into the *K*-level, and the K_α-line is emitted in this way. A transition of an electron from the *M*-level into the *K*-level is accompanied by the emission of the line K_β, whereas the L_α-line is emitted by the transition of an electron from the *M*-level into the *L*-level.

Experimentally there are two methods of determining the energy difference between the level corresponding to the normal state and the

K-level. One of them consists in the determination of the excitation potential V_0 of the *K*-series, from which we obtain the smallest kinetic energy that the colliding electron must have in order to remove an electron from the *K*-group; and the other is based on the determination of the 'limiting frequency' of the *K*-absorption jump or absorption edge which just suffices, according to Einstein's relation $eV_0 = h\nu$, to impart to the atom during absorption the same amount of energy as is possessed by an electron of velocity V_0 volts. (For information on the number of levels and lines cf. p. 63.)

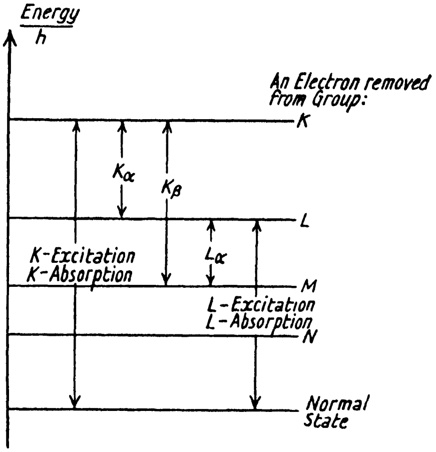

FIG. 33. Production of the X-ray Spectrum.

4. Quantum Mechanics and Wave Mechanics

It is necessary to differentiate between nuclear properties (mass of the ionized atom, radioactivity, etc.) and peripheral or electron properties of the atom; the chemical behaviour of the elements is determined by the number and strength of binding of the outer electrons in their atoms; in contrast to optical spectral lines, X-ray lines are due to quantum jumps in the more deeply seated levels of the electronic shell of the atom. These and other fundamental facts were responsible for the introduction of the model of the atom already described and went hand in hand with its development. In spite of these facts, however, it was later necessary to replace the atomic model by conceptions of a less concrete nature, first, in order to be able to interpret more complicated spectra, for the mathematical treatment of which the former conception had failed, and, secondly, in order to avoid difficulties of an epistemological nature. The advance took place along two different lines, namely, quantum mechanics and wave mechanics. In quantum mechanics the course followed was one indicated by the principle of correspondence, whereas wave mechanics was developed from the initial conception that the behaviour of electrons must be described in a dualistic manner. In the latter case, not only the corpuscular nature of electrons comes into evidence, but they also possess properties that can only be grasped by their theoretical treatment as waves (material waves).

The principle of correspondence may be stated as follows: The energy changes in the atom do not take place continuously, but stepwise. The development of the steps is very marked when the energy changes take place between low quantum states, and as a rule the emission of short waves with large values of $h\nu$ is associated with these changes; but when we pass over to higher and higher quantum states, with a corresponding diminution of $h\nu$, the development of the steps degenerates in increasingly greater measure. Complete degeneration of the steps, however, is synonymous with a transition from the quantum-theoretical to the classical domain, in which the processes do not occur in steps, but in a continuous manner. Classical physics is thus revealed as a limiting case of quantum physics.

In quantum mechanics we refrain from stating the position and velocity of the electrons in the atom. Instead of this we make use of directly observable quantities for the characterization of the states of the atom, to wit, the frequencies and intensities of spectral lines (virtual resonators), as well as the energy levels of atoms as established by experiments on electron collisions. The relations obtained in this way embrace both the processes to be described by the quantum theory, in which the quantum of action unavoidably occurs, and also the processes to be described by classical theory, in which the existence of a quantum of action cannot be observed. The wave-mechanical viewpoint is rendered more intelligible by reference to the theory of light.

Light, in addition to having wave properties which are revealed say in the phenomena of diffraction, has also corpuscular properties which manifest themselves in the photo-electric effect, where light quanta or photons are the effective agents. In the case of electrons the sequence is reversed, for wave mechanics ascribes to them not only corpuscular properties, but also wave properties. From this point of view matter, or the electrons and protons composing it, appears resolved into waves. In wave mechanics a vibration law is formulated for the atom which is quite similar to the law of the mechanical vibrations of strings. But here we are not dealing with the motion of material particles, but with that of an abstract quantity called the Schrödinger ψ-function, which is only mathematically intelligible. The probability that an electron will be situated in a particular position at a definite time is proportional to the square of the magnitude of ψ in this region. If ψ is restricted for the most part to a single region (as say in the case of an emitted electron), we can assume with a probability bordering on certainty that the electron will also be found in this confined region. But when we are dealing with an electron in the atom, e.g. the electron in the hydrogen atom, the ψ-waves are distributed over the whole atom, and the electron cannot be localized at a definite place in the atom, but its presence will be more probable at certain places than at others.

The wave-mechanical viewpoint is also of great importance in the study of radioactivity, for it makes the process of disintegration more intelligible, as indicated on p. 98.

5. Mathematical Appendix to the Chapter on Atomic Structure

Let us consider the motion of an electron and of a positive nucleus, the mass of which relative to that of the electron can be assumed to be infinitely large, and for purposes of simplicity let us restrict ourselves to circular orbits.

By Coulomb's law the force of attraction between the nucleus of charge Ne and an electron of charge e amounts to $\frac{Ne^2}{a^2}$, whilst the equal but oppositely directed centrifugal force has the value $\frac{mv^2}{a}$, where m denotes the mass of the electron, v its velocity, and a the radius of the electron orbit. Such an atom, built up after the manner of a planetary system, will hold together when

$$\frac{Ne^2}{a^2} = \frac{mv^2}{a}. \tag{1}$$

The theory of series spectra requires that the angular momentum mva of the electron in the stationary orbits cannot assume arbitrary values, but must always be an integral multiple of $\frac{h}{2\pi}$, i.e.

$$mva = \frac{nh}{2\pi}, \tag{2}$$

where n is a whole number.

By combining equation (2) with equation (1) we can calculate the radius of the orbit of the electron, its velocity, and also the work (W) necessary to separate it completely from the atom, or to remove it to infinity, i.e. to 'ionize' the atom. Moreover, we can also calculate from this the revolution frequency (ω) of the electron in its stationary orbit. We thus obtain:

$$a = \frac{n^2h^2}{4\pi^2Ne^2m}, \tag{3}$$

$$v = \frac{2\pi e^2 N}{hn}, \tag{4}$$

$$W = -\tfrac{1}{2}mv^2 + \frac{Ne^2}{a} = \frac{2\pi^2 e^4 m N^2}{h^2 n^2}, \tag{5}$$

$$\omega = \frac{4\pi^2 m e^4 N^2}{h^3 n^3}. \tag{6}$$

On the lines of the theory described, the emission of spectral lines takes place when the electron jumps from one of the orbits permissible by the quantum hypothesis into an orbit of lesser energy situated nearer the nucleus, e.g. from the n_1th orbit to the n_2th orbit. If ν denote the frequency of the spectral line, and W_n the energy content of the system in the state n, then

$$\nu h = W_{n_2} - W_{n_1}. \tag{7}$$

For an arbitrary atom we have

$$\nu = \nu_0 N^2\left(\frac{1}{n_2^2} - \frac{1}{n_1^2}\right), \tag{8}$$

where $\nu_0 = \frac{2\pi^2 e^4 m}{h^3} = 109{,}737$ (Rydberg's constant) $\times 3 \times 10^{10}$, and N is the atomic number of the element. For the frequency of the hydrogen lines (Balmer series) we obtain

$$\nu = \nu_0\left(\frac{1}{n_2^2} - \frac{1}{n_1^2}\right) = (n_1 - n_2).\nu_0.\frac{n_1 + n_2}{n_1^2 n_2^2}. \tag{9}$$

When n_1 and n_2 are large compared with their difference we can put $n_1 = n_2 = n$ in the last factor, and by taking account of equation (6) we obtain for the frequency

$$\nu = (n_1 - n_2) \times \frac{2\nu_0}{n^3} = (n_1 - n_2)\omega.$$

From this we see that there exists in the region of large values of n a quantitative relation between the revolution frequency and the frequency of the radiation, i.e. a 'correspondence' between the latter and the motion of the electrons in the atom in the region of large quantum numbers. The harmonics in the radiation corresponding to $(n_1 - n_2) > 1$, however, are only present when the motion of the electron is elliptical, and not when it is circular.

If we associate a material wave with every particle, then the connexion between the frequency of this wave (ν) and the mass (m) is given by $h\nu = mc^2$, where c = the velocity of light, and the velocity of propagation of this wave is $\mu = \lambda\nu$. Since the product of the mechanical velocity

(v) of the particle and the velocity of propagation (μ) of its associated wave $\mu v = c^2$, we obtain

$$\lambda = \frac{c^2}{v\nu},$$

and by taking account of the first equation, we deduce

$$\lambda = \frac{h}{mv}.$$

The de Broglie wave-length λ of electrons with a velocity of about one-third that of light is of approximate magnitude 10^{-9} cm., and of slow electrons such as are emitted by luminous heated wires about 10^{-7} cm.

The fundamental equation of wave mechanics (Schrödinger's differential equation) is:

$$\Delta\psi + \frac{8\pi^2 m}{h^2}(E-V)\psi = 0,$$

where E denotes the total energy and V the potential energy of the particle.

A single-valued, finite, and continuous solution of the differential equation is found only for particular values of the parameter E which occurs in the equation, and these are called the 'proper'- or 'eigen'-values of the differential equation. The evaluation of the eigen-values of the differential equation thus supplies the required quantum energy-values. In this way we obtain for the energy of a linear harmonic oscillator, in agreement with experimental evidence,

$$E = (n+\tfrac{1}{2})h\nu.$$

The energy is accordingly always an odd multiple of half an energy-element $\left(\frac{h\nu}{2}\right)$, whereas this energy should be an integral multiple of a whole energy-element ($h\nu$) from the viewpoint of considerations based on atomic models. The series of possible energy-steps is thus $\frac{h\nu}{2}$, $3\frac{h\nu}{2}$, $5\frac{h\nu}{2}$, etc., whilst on the earlier mode of consideration the series of steps should be 0, $h\nu$, $2h\nu$, and so on.

IX

THE ATOMIC NUCLEUS

1. The Constituents of the Nucleus

As discussed in Chapter VIII, it follows from the large deflexion of individual α-particles from their rectilinear path in their passage through atoms, that the positive charge responsible for their deviation is situated at the centre of the atom, and that the atomic nucleus, which is the carrier of this charge, occupies an extremely small space. The atomic nucleus contains the positively charged α-particles which leave the atom in the process of disintegration. β-particles are also emitted by the nucleus. This follows from the fact (p. 154) that the extensive chemical and spectral changes experienced by the atom in consequence of the emission of an α-particle ($++$) are entirely annulled by the loss of two β-particles ($--$), a phenomenon that admits of no other explanation than that the α- and β-particles are expelled from the same part or the nucleus of the atom. It should be noted, however, that in the process of atomic disintegration only the primary β-particles originate directly from the nucleus. The secondary β-particles, which are produced during the passage of primary β-, as well as α-, γ-, or other rays through the atoms, originate in the electron cloud surrounding the nucleus, just like the electrons that are involved in the phenomenon of ionization or in other chemical processes. Thus the β-rays that are emitted along with the α-rays from radium, radiothorium, and radioactinium have their origin exclusively in the electron shell of the atoms (see below).

The α-particles, however, are not the ultimate bricks of which the nucleus is composed, for in the process of the artificial disruption of atomic nuclei protons, and in other cases neutrons, are ejected from the nuclei. We must regard these protons and neutrons as the ultimate constituents of the nuclei, and consider the α-particle as a kind of 'molecule' of the 'nuclear atoms' possessing especial stability, and arising from the union of 2 protons and 2 neutrons. If we make this assumption, we need no longer assume that electrons are present in the nucleus; the nuclear or primary β-particles that we observe are produced immediately before their emission by the transformation: neutron $=$ proton$+\beta$. The atomic number expresses the number of protons in the nucleus, whereas the number of neutrons in a particular nucleus is obtained by subtracting the atomic number from the integer nearest the atomic weight. Isotopes accordingly differ in the number of neutrons contained in their nuclei, the simplest case being that of deuterium, the nucleus of which consists of a single proton in association with a neutron. Even though α-particles are not to be regarded as the ultimate constituents of the nucleus, they play a very important part in its structure. In the spontaneous nuclear decay of radioactive substances α-particles are frequently ejected, but never protons, and for nuclei whose mass is a multiple of 4 we never observe disruption when they are subjected to the influence of α-particle bombardment. The great stability of the α-particle is explained by the comparatively large loss in mass entailed in its formation from protons and neutrons, a process which is of a strongly exothermal nature.

The most direct method of evaluating the nuclear charge (see p. 32) of an element is to measure the scattering which α-particles experience when they pass through its atoms; but the most convenient way is the calculation of the nuclear charge from the wave-length of characteristic X-ray lines on the basis of Moseley's relation (see p. 63).

Information on nuclear structure is obtained mainly from the study of the following: (*a*) the scattering of α-particles (and in part also of β-particles) in their passage through atoms; (*b*) α-particles of long range and the fine structure of the α-rays; (*c*) the wave-lengths of the penetrating γ-rays which leave the atom in the disintegration process; (*d*) the disruption of the atom; (*e*) artificial radioactivity; (*f*) the deviation from integers of the mass of the isotopic types of atom; (*g*) the cosmical frequency distribution of the elements.

2. The Law of Force in the Neighbourhood of the Nucleus

In the investigation of the scattering of α-particles (cf. also the discussion on p. 31 et seq.) a strong pencil of α-particles of definite velocity is allowed to strike a thin sheet of the scattering substance, and by means of scintillation counts the number of particles deflected through say 135° is determined. As a rule, for every 10^5 incident particles about one deflected particle is observed. By variation of the velocity of the incident particles the dependence of the scattering on the energy of the particles can be determined. If the electrostatic repulsion between the positive nucleus and the positive α-particle is consistent with Coulomb's law, it is to be expected that the number of scattered α-particles will be inversely proportional to the square of their energy. The scattering of α-particles by atoms of the elements which lie between uranium (92) and copper (29) is found to be in accord with this expectation.

The nearest distance of approach of an α-particle to the nucleus of say copper cannot be smaller than the radius of the copper nucleus, for otherwise the radius of the copper nucleus would have an influence on the angular distribution of the deflected α-particles. Thus the distance of closest approach of an α-particle to the nucleus yields the upper limit for the radius of the copper nucleus, and this is found to be about 10^{-12} cm. If we consider the case of a uranium nucleus instead of a copper nucleus and use the fastest α-particles, we find for the distance of nearest approach of the α-particle to the nucleus and hence for the upper limit of the radius of the uranium nucleus the value 3×10^{-12} cm.

Should the α-particle penetrate into the nucleus of the atom, the validity of Coulomb's law must cease and in place of it another law will be effective. The occurrence of a different law of force from that of Coulomb in the immediate neighbourhood of the nucleus is observed in the investigation of the phenomenon of the scattering of α-particles by lighter atoms such as magnesium or aluminium. For very slow α-particles we find here also only a small deviation in the number of scattered particles from the theoretical value calculated on the basis of the validity of Coulomb's law; but when the velocity of the α-particles is increased we find at first a marked diminution and then an increase in the number of scintillations observed experimentally, and the number observed is always less than that calculated theoretically. In Fig. 34 the ordinates represent the numbers of scintillations counted, expressed as percentages of the values calculated theoretically, whereas the abscissae represent the reciprocal values of the energy of the incident α-particles, for the case of aluminium and for a scattering angle of 135°. The broken line represents the results for gold as the scattering substance, and here the value established experimentally amounts to 100 per cent. of the value calculated, since Coulomb's law of force holds strictly for the approach of α-particles to heavy nuclei.

The anomalous scattering shown in Fig. 34 for the case of aluminium and other light atoms must be interpreted by assuming that for close approach to the lighter nuclei a force of attraction is superposed on the Coulomb force of repulsion, the observed effects being due to the force resultant from these two components.

Apart from that described above, there is yet a second method of calculating the magnitude of the nuclear radius, to wit, from the velocity of the α-particles emitted. If we assume that an α-particle in its expulsion from the nucleus is released from the surface with zero velocity and is then repelled in accordance with Coulomb's law, whereby both the α-particle and the residual nucleus are to be regarded as point charges, we can then calculate the required nuclear radius in a simple manner (law of potential) from the energy of the α-particle observed experimentally.† For the uranium nucleus we obtain in this way $r \geqslant 6{\cdot}5 \times 10^{-12}$ cm., whereas the above-mentioned deflexion experiments

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† The energy $E = \frac{2e(Z-2)e}{r}$, where r = nuclear radius, e = elementary electron charge, Z = atomic number.

yielded the quite different value $r \leqslant 3{\cdot}5\times 10^{-12}$ cm. Thus when we compare the behaviour of α-particles in the process of scattering by the nucleus with that when they escape from the nucleus, we are led to conflicting results. This contradiction manifests itself also in another form, which we shall consider in what follows. In order to explain the phenomenon of scattering we must assume the existence both of a repulsive and an attractive force, of which the former predominates at larger distances, and the latter at smaller distances from the centre of the nucleus. If we plot the potential energy of an α-particle in its dependence on the distance from the centre of the nucleus, we obtain Fig. 35. The maximum of the curve corresponds to the distance from the centre of the nucleus, for which the attractive and repulsive forces just balance one another. Within this distance the attractive forces are the determining factor and the potential can assume a negative value, whereas at greater distances from the nucleus the potential due to the electrostatic forces diminishes with increasing distance. On the basis of classical dynamics we can calculate the potential at a distance of 5×10^{-12} cm. from the centre of the nucleus, and for uranium this works out to be about 5 million volts. Since the fast α-particles possess energy of this magnitude, they should be in a position to penetrate into the nucleus of the uranium atom. The phenomenon of scattering, however, shows that this is not the case, and that α-particles are prevented from penetrating into the nucleus. It is possible to eliminate the contradictions mentioned if we turn our attention to the wave-mechanical mode of treatment described on p. 89 et seq. Considerations that have been

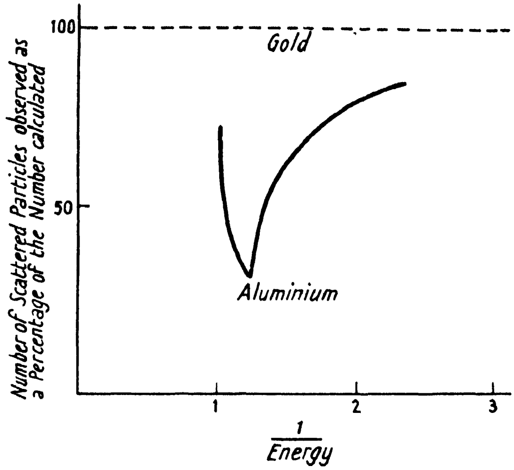

FIG. 34. Deviation from Coulomb's Law near the Nucleus. Scattering of α-Particles through 135° by Aluminium and Gold.

undertaken on this basis lead to a curve whose maximum lies much nearer to the nucleus than that of Fig. 35 ($0{\cdot}7\times10^{-12}$ cm. instead of 5×10^{-12} cm.). Correspondingly, the maximum protective potential lies very much higher, at 30 million volts instead of at 5 million volts, and falls off very rapidly in the direction of the nucleus. The nucleus is thus protected by a barrier at an exceedingly high potential. On the basis of classical ideas no α-particle should be able to surmount this barrier in either direction, and if α-particles are emitted from the nucleus at all, they ought to escape with a markedly higher energy (about 30×10^6 volts) than that found from experiment. However, if we make use of the ideas of wave mechanics, it is possible for an α-particle, or the wave system with which we identify it, occasionally to vacate the nucleus, without having to surmount the potential barrier. From the viewpoint of classical mechanics the particle will never be able to escape (i.e. surmount the barrier); wave mechanics, on the other hand, concedes a finite probability of escape through the thin barrier. The greater the energy of the α-particle and the thinner and lower the barrier through which it passes, the greater will be the probability of its escape. In this way arises a close connexion between the energy of an α-particle and the decay period of the element emitting it. This is the relation of Geiger and Nuttall, which we shall discuss on pp. 105 and 146.

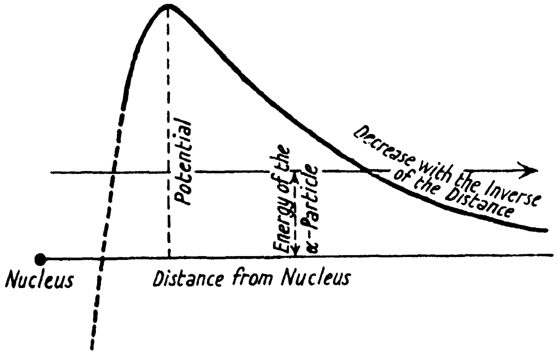

FIG. 35. Variation with Distance of the Potential Energy of an α-Particle in the Field of the Uranium Nucleus.

3. Energy Levels of Atomic Nuclei

We have seen on p. 80 et seq. how the idea of energy levels was applied with the greatest success in the explanation of the structure of the extra-nuclear part of the atom. In what follows we shall discuss the considerations that have led us to assume the existence of energy levels also in the atomic nucleus, and the nuclear energy level schemes that have been obtained.

One of the most important observations which led to the assumption of energy levels in the nucleus was the result that not all the α-particles that are emitted by the nuclei of certain radio-elements have the same velocity, as in the case of the α-particles from ThC or AcC (see p. 26). For these individual radio-elements other sharply defined lines occur along with the main line in the velocity spectrum of the α-particles emitted by them. Thus the following five groups of α-rays are found in the α-ray spectrum of ThC (Table 24). From the figures entered in the table we see that of the α-rays emitted in the transformation of ThC into ThC″ only part of them are ejected with the maximum energy of $6{\cdot}2 \times 10^6$ e.V., the remaining groups having a smaller energy content. The difference between these latter energy values and $6{\cdot}2 \times 10^6$ e.V. is retained in the resulting ThC″ nuclei. Accordingly, some of the ThC″ nuclei have a surplus content of energy, so that from the viewpoint of energy levels they are in an excited state. From the fact that there are four different groups of α-particles with a smaller energy content than the maximum we can conclude that there must be four different levels of excitation in the nucleus of ThC″. There will be an additional fifth level with a normal energy content, and this will exist in all ThC″ nuclei which have been produced from ThC after the emission of an α-particle of energy $= 6{\cdot}2 \times 10^6$ e.V.

TABLE 24

Energies of the α-Ray Groups emitted by ThC

Group	*Energy in e.V.*
α_1	$6{\cdot}201 \times 10^6$
α_2	$6{\cdot}161$,,
α_3	$5{\cdot}873$,,
α_4	$5{\cdot}729$,,
α_5	$5{\cdot}709$,,

Excited states have only a limited life, and we should expect that the energy surplus of the excited nuclei will gradually be emitted in the form of γ-rays, in a manner analogous to the production of spectral lines in optical spectroscopy (p. 81). This is just what happens, for ThC″ emits γ-rays, and in the spectrum of these rays we find a series of lines with the frequencies to be expected. Thus from the results of the above table we should expect an amount of energy given by $6{\cdot}201 - 6{\cdot}161 = 0{\cdot}040$ million e.V. to be liberated in the transition from level I of the ThC″ nucleus to the ground level (level 0); this amount of energy does actually appear in the form of γ-rays in the spectrum of the γ-rays emitted by ThC″. Likewise in the transition from level II to level I, and so on, in the nucleus of the ThC″ atom, the quantities of energy liberated reappear in the form of lines in the resulting γ-ray spectrum, as may be seen from Table 25.

TABLE 25

Energy Differences between the Groups of α-Rays emitted by ThC, and Energies of the Lines in the γ-Ray Spectrum of ThC″ (Energies in 10^6 e.V.)

Transition between levels	*Energy difference of the α-ray groups*	*Energy of the γ-line*
$L_I - L_O$	0·040	0·040
$L_{II} - L_O$	0·328	0·327
$L_{III} - L_O$	0·472	0·471
$L_{II} - L_I$	0·288	0·287
$L_{III} - L_I$	0·432	0·432
$L_{IV} - L_I$	0·452	0·451

Also in the case of a β-ray disintegration it is very frequently found that the resulting nucleus is in an excited state, and in most cases the energy surplus contained in the excited nucleus is radiated in the form of γ-rays, the nucleus passing over into the normal state. Not until this state has been reached, when we are dealing with a nucleus that emits α-rays, does the emission of an α-particle occur. Very infrequently, however, we are presented with another picture. Here the nuclei which are left in a highly excited state after the emission of β-particles do not first emit γ-quanta, but the energy of excitation is added to the energy of the normal α-particles, so that more energetic α-particles of longer range are emitted in place of those of normal range. Thus the normal α-particles from RaC′, which is produced by β-disintegration from RaC, have an energy of $7{\cdot}68 \times 10^6$ e.V., but the emission of every 10^6 particles of normal range is accompanied by the emission of about 22 particles of long range. A more detailed investigation has shown that these particles of long range are distributed among some 12 groups of different energy content. Now we have already seen that the energy of the α-particles of long range is due to the fact that the energies of excitation of the nuclei are added to the energy of the normal α-particles, which we may regard as being emitted from the normal or ground level of the nucleus. In view of the fact that there are thus 13 velocity groups of α-particles, we can deduce the initial existence of at least 13 energy levels in the nucleus of RaC′. In the γ-spectrum of RaC′ there are a number of lines which correspond to the differences of many of these energy levels, and this can be regarded as a further argument in favour of the existence of such levels. In order to be able to interpret all the lines in the γ-ray spectrum we must assume the existence of two additional levels in the nucleus of RaC′. These are the considerations which have led to the scheme of nuclear levels represented in Fig. 36. The mass number and atomic number of the relevant nucleus are given for each system of levels, as well as the radioactive disintegration to which the succeeding nucleus owes its excitation. Nuclei of different mass number are separated by vertical lines.

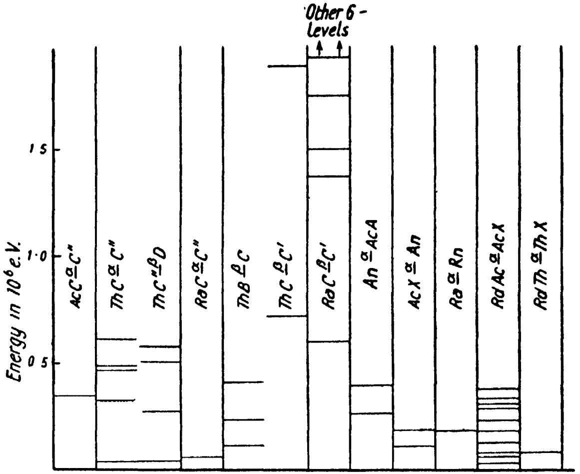

FIG. 36. Scheme of Energy Levels of different Nuclei, obtained by the combined Analysis of the Fine Structure of α-Rays, of the Long-Range α-Rays, and of the γ-Rays.

The cases discussed above have reference to nuclei whose state of excitation is low. Under the action of neutrons it is possible to obtain high nuclear energy levels. When a neutron is taken up by a nucleus a very large amount of energy is liberated, to wit, the energy of binding of the neutron, which usually amounts to some 7 to 8×10^6 e.V. This energy at first appears as energy of excitation of the newly created nucleus, and is then gradually emitted in the form of γ-radiation. Experiment has revealed an extraordinarily large number of nuclear levels in this energy region; they are accounted for by the large number of nuclear components (neutrons and protons) involved in the building up of the heavier nuclei [1].

4. Origin of the β- and γ-Radiations

In the disintegration of substances with emission of β-rays each disintegrating atom emits one β-particle. The emission of γ-rays is a phenomenon associated both with β- and with α-ray disintegration. When the γ-rays pass through the electron shell of the atom they release secondary β-particles, so that these do not originate in the nucleus. In most cases the number of secondary β-particles exceeds the number of primary β-particles, which have their origin in the nucleus. By the measurement of the deflexion of β-rays in a magnetic field, line spectra of the rays have been observed, but they are of secondary origin. One way of showing this is to allow γ-rays from various radio-elements to fall on different substances and to investigate in a magnetic field the energies of the secondary β-rays liberated from the substances. By this means it is possible to produce in foreign atoms the magnetic line spectra which otherwise are produced in the shells of the atoms which emit the β-rays. Substances like radium, radiothorium, and radioactinium are α-rayers, but in spite of this they exhibit a magnetic β-spectrum. This fact supplies us with the most direct proof that the magnetic β-spectrum is of secondary origin.

The connexion between the energy of the γ-impulse and that of the β-particles ejected from the electron groups of the atom is as follows: The energy of the ejected β-particle is equal to the energy ($h\nu$) of the γ-impulse less the work which must be performed in order to release the electron from the electron group involved in the process. This work will be different for the various groups from which the electron may be released. Let us consider, for example, the very simple spectrum of radium. There are three β-lines present in this spectrum and their energy values are $0{\cdot}876\times10^5$, $1{\cdot}720\times10^5$, and $1{\cdot}856\times10^5$ e.V. The first two differ in energy by $0{\cdot}844\times10^5$ e.V., and the last two by $0{\cdot}136\times10^5$ e.V. From these values it follows that the γ-quanta which leave the nuclei liberate secondary electrons from the K-, L_{I}-, and M_{I}-levels of the radon atoms resulting from the disintegration of atoms of radium, since $0{\cdot}844\times10^5$ is almost equal to the difference between the works of separation ($0{\cdot}981\times10^5$ and $0{\cdot}180\times10^5$ e.V.) of a K- and of an L_{I}-electron, whereas $0{\cdot}136\times10^5$ e.V. is the difference between the works of separation for an L_{I}-electron and for an M_{I}-electron ($0{\cdot}045\times10^5$ e.V.), all for an atom of radon. The values for the works of separation are known from X-ray spectroscopic data (cf. p. 59). The importance of these results lies in the fact that as soon as we know the energy of the β-ray produced and the work necessary to separate it from its particular level, we can immediately obtain the energy of the γ-ray emitted by the nucleus, which is simply the sum of these two quantities. In some cases this is the only method of evaluating the energy of the γ-ray. The value of the energy of a γ-ray from radium is $1{\cdot}89\times10^5$ e.V. In general it is necessary to assume that several different γ-rays are emitted from the nuclei of a radioactive substance, in order to be able to interpret the lines of the secondary β-spectrum. It will be seen from Table 26 that the γ-spectrum of RaC consists of 15 lines.

It was at one time assumed that the γ-ray in its passage through the electron shell gives rise to photo-electric effects, and in this way produces the discrete groups of β-rays. But it has been shown that the intensity of the β-ray groups is much too large to be explained by an

TABLE 26

γ-Ray Lines of RaC (83)

Energy of the γ-line	*Levels from which the β-rays originate*	λ (*in X.U.*)	*Assumed works of separation of an electron for the element* 84 (*e.V.*)
$0{\cdot}589\times10^5$	L_I	209·5	
$2{\cdot}75\times10^5$	$K\ L_I$	44·9	$K = 0{\cdot}928\times10^5$
$3{\cdot}32\times10^5$	$K\ L_I$	37·2	
$3{\cdot}89\times10^5$	$K\ L_I$	31·7	$L_I = 0{\cdot}168\times10^5$
$4{\cdot}29\times10^5$	$K\ L_I$	28·8	
$5{\cdot}03\times10^5$	K	24·5	$L_{II} = 0{\cdot}161\times10^5$
$6{\cdot}12\times10^5$	$K\ L_I\ M_I\ N_I$	20·2	
$7{\cdot}73\times10^5$	K	16·0	$L_{III} = 0{\cdot}135\times10^5$
$9{\cdot}41\times10^5$	K	13·1	
$11{\cdot}30\times10^5$	$K\ L_I\ M_I\ N_I$	10·92	$M_I = 0{\cdot}041\times10^5$
$12{\cdot}48\times10^5$	$K\ L_I$	9·89	
$13{\cdot}90\times10^5$	K	8·89	
$14{\cdot}26\times10^5$	$K\ L_I\ M_I$	8·66	
$17{\cdot}78\times10^5$	$K\ L_I$	6·94	
$22{\cdot}19\times10^5$	$K\ L_I$	5·57	

ordinary photo-electric effect, and in many cases there is a very rich yield of electrons liberated from the shell of the atom by the γ-rays. Thus in the case of ThB the γ-line of wave-length 51·2 X.U. produces a yield of electrons from the K-level of more than 25 per cent., and in the case of the γ-radiation from RaD a separation yield of more than 80 per cent. has been established for all the levels. The γ-lines that possess most energy are those of energy $26{\cdot}5\times10^5$ and $22{\cdot}2\times10^5$ e.V., emitted respectively from ThC″ and RaC, whereas the softest γ-lines are emitted by RdAc and ThC″ with respective energy values $0{\cdot}32\times10^5$ and $0{\cdot}41\times10^5$ e.V., as obtained from the magnetic spectrum of the rays.

The energy emitted in the form of γ-radiation in the process of nuclear disintegration can be determined either calorimetrically or by the measurement of the ionization produced by the γ-rays. The amount of heat developed per hour by 1 gm. of radium is 25·5 calories, and of this 0·95 calories are to be attributed to the γ-radiation emitted by radium, the effect of the total secondary β-radiation being included in the latter figure. In the calorimetric method of determining the γ-ray energy the following points should be noted: (*a*) the effect of the α-radiation predominates; (*b*) the γ-rays are very penetrating; and (*c*) it is difficult to separate the effect of the γ-radiation from that of the β-radiation. For these reasons the calorimetric method is less satisfactory than the ionization method. Thus it is found that the γ-rays from an amount of Ra(B+C) equivalent to 1 gm. of radium produce $2{\cdot}2\times10^{15}$ pairs of ions per second in air, and since the energy required to produce a pair of ions in air is 32·5 e.V., it follows that the quantity of energy radiated in the form of γ-rays from this amount of Ra(B+C) is 8·8 calories per hour. The mean value of the energy emitted in the form of γ-radiation during the disintegration of a RaB atom together with the succeeding product is $1{\cdot}86\times10^6$ e.V.

5. The Continuous Spectrum of β-Rays

The β-rays emitted by one and the same radio-element, the nuclei of which disintegrate by ejection of β-particles, have velocities that vary over a wide range. We are here confronted with a continuous spectrum, in contrast to the line spectrum shown by the resolution in a magnetic field of β-particles liberated from the atomic shells. If we disregard its very feeble γ-radiation, RaE emits only β-rays, and is a particularly suitable substance for the investigation of the continuous β-ray spectrum. This can be carried out either by experiments on the magnetic deflexion of the rays or by absorption measurements. It is found that the energy content of the β-particles ejected by the nucleus of RaE varies between $1{\cdot}23\times10^6$ e.V. and 2×10^5 e.V., so that the electrons that leave the nucleus of RaE differ in their energy values by more than 1×10^6 e.V. By the measurement of the development of heat by the whole of the β-radiation from RaE it has been possible to show that these energy differences do not arise from the occurrence of say secondary processes connected with the passage through the atomic shells of the β-particles coming from the nucleus. This development of heat corresponded to $3{\cdot}37\times10^5$ e.V. per particle, and was equal to the mean energy of the continuous β-spectrum (cf. Table 27). Thus the transference of nuclear β-rays into secondary radiation does not take place in the outer shell of the RaE atom, and the various RaE nuclei emit β-particles of different energy values. If the half-value period of freshly deposited RaE be determined and compared with that for an older preparation, most of which has already decayed, the two values of the period are found to be accurately identical, and the same statement holds in the case of the succeeding disintegration product RaF. From this it follows that the energy content of all the RaE atoms must be the same, and likewise in the case of all the atoms of RaF. This is an unexpected result, since the various RaE atoms disintegrate into RaF under emission of different amounts of β-ray energy.

In order to resolve this contradiction it is assumed that, simultaneously with the emission of a β-particle, every RaE nucleus also emits a second very light particle that does not ionize. The energy of this 'neutrino', together with that of the electron (β-ray), is assumed to be equal to the difference in energy content between the original nucleus and that of the succeeding product of disintegration. Thus the sum of the energies of the electron and the neutrino is the true energy of disintegration. It can be obtained from the upper limit of the β-spectrum; for these fastest electrons the energy of the neutrino is zero. According to this hypothesis, the emission of β-radiation is not a unitary elementary process like the emission of a light quantum, but a dual process consisting of the simultaneous emission of an electron and a neutrino with predetermined total energy.

The energy corresponding to the upper limit of the continuous β-spectrum for several radio-elements is shown in Table 27, which also contains the values of the average energy of the β-spectrum in each case, and the half-value periods of the radio-elements concerned.

TABLE 27

Continuous β-Ray Spectrum of several Radio-elements

Radio-element	*T*	*Average energy (in 10^5 e.V.)*	*Energy of the upper limit (in 10^5 e.V.)*
UX_2	1·14 min.	8·2	23·2
RaB	26·8 „	2·3	6·5
RaC	19·7 „	7·4	31·5
RaE	5·0 days	3·4	12·3
$MsTh_2$	6·13 hours	5·6	22·5
ThB	10·6 „	0·89	3·6
ThC	60·5 min.	8·0	22·0
ThC″	3·1 „	5·8	18·0

6. Regularities within the Disintegration Series

There is a simple relation between the disintegration constants of the radio-elements and the ranges of the α-particles emitted. If, as in Fig. 37, we plot the logarithm of the range as abscissa and the logarithm of the disintegration constant as ordinate, we obtain for each of the three transformation series a separate straight line. The relation fails in individual cases, as in those of RaC′ or AcX. However, the formula† calculated on the basis of wave mechanics can be applied with some success even in the last-named cases. In addition to the quantities mentioned, this formula also involves the atomic number and the nuclear radius.

A similar relation to that which holds for α-rays is also obtained for β-rays, if we plot the logarithm of the disintegration constant against the logarithm of the energy of the upper limit of the continuous β-ray spectrum (Fig. 38). It will be seen that the points fall on two distinct curves. In explanation of this, the view has been advanced that we must distinguish between those radio-elements whose disintegration leads to a nucleus with the same angular momentum as the disintegrating nucleus, and those for which this is not the case. The upper graph in

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† See footnote on p. 146.

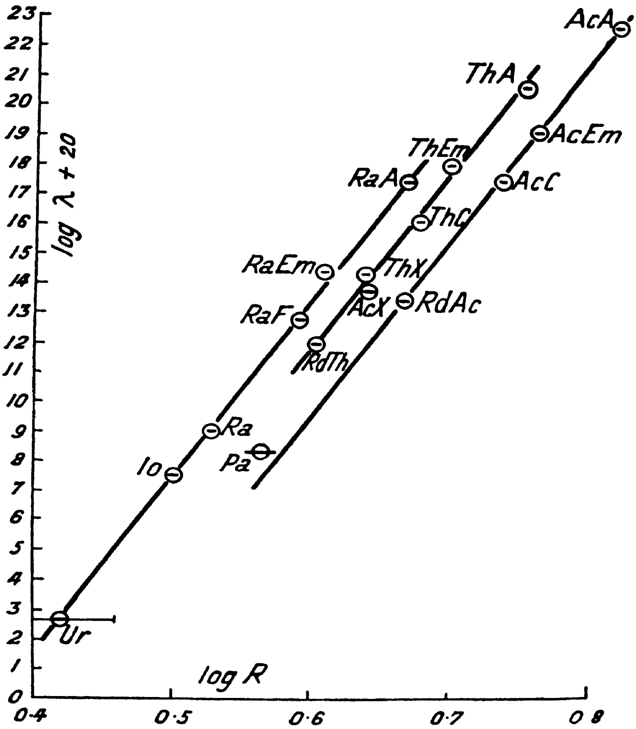

FIG. 37. Relation between the Range and the Disintegration Constant for α-Rayers.

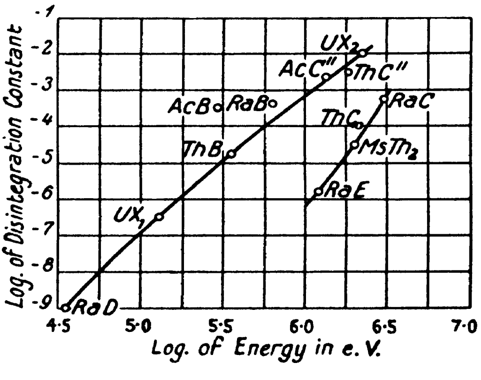

FIG. 38. Relation between the Energy of Disintegration and the Disintegration Constant for β-Rayers.

Fig. 38 embodies the results for radio-elements whose nuclear angular momentum does not differ from that of the nucleus resulting from the β-ray disintegration, whereas the lower graph refers to radio-elements for which the β-ray disintegration is accompanied by an alteration of one unit in the angular momentum. In general, the probability of disintegration increases when there is no change in the angular momentum after disintegration.

REFERENCES

A detailed description of the properties of atomic nuclei is to be found in *Structure of Atomic Nuclei and Nuclear Transformations*, by G. GAMOW (Oxford University Press, 1937). The following books will also be found useful in connexion with Chapters IX and X. *An Introduction to Nuclear Physics*, by N. FEATHER (Cambridge University Press, 1936); *Elements of Nuclear Physics*, by F. RASETTI (Blackie, London, 1937); *Einführung in die Kernphysik*, by H. KALLMAN (Deuticke, Vienna, 1937); *Die Atomkerne*, by C. F. von WEIZSÄCKER (Akad. Verlagsgesellschaft, Leipzig, 1937).

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X

THE TRANSMUTATION OF THE ELEMENTS

THE elements owe their great stability to the protection afforded to the atomic nucleus by the potential barrier arising from the powerful electric field surrounding it. Charged particles, rich in energy, are able to break through this barrier and effect a transmutation of the atomic nucleus, but the harvest is in general a very modest one. As a rule the particle which succeeds in penetrating the barrier is taken over by the nucleus, and in many cases this process leads to the ejection of a proton or another heavy particle. The outcome of all this is the formation either of a stable nucleus or of a radioactive unstable nucleus, in which latter case we are presented with an instance of radioactivity produced by artificial means. Let us first discuss the various types of nuclear transmutation without regard to whether or not the resulting nucleus is stable.

The following *types of nuclear transmutation* are known:

(*a*) ${}^{14}_{7}\mathrm{N} + {}^{4}_{2}\alpha = {}^{17}_{8}\mathrm{O} + {}^{1}_{1}\mathrm{H}$
(*b*) ${}^{9}_{4}\mathrm{Be} + {}^{4}_{2}\alpha = {}^{12}_{6}\mathrm{C} + {}^{1}_{0}n$
(*c*) ${}^{9}_{4}\mathrm{Be} + {}^{1}_{1}\mathrm{H} = {}^{6}_{3}\mathrm{Li} + {}^{4}_{2}\mathrm{He}$
(*d*) ${}^{9}_{4}\mathrm{Be} + {}^{1}_{1}\mathrm{H} = {}^{8}_{4}\mathrm{Be} + {}^{2}_{1}\mathrm{D}$
(*e*) ${}^{7}_{3}\mathrm{Li} + {}^{1}_{1}\mathrm{H} = {}^{8}_{4}\mathrm{Be}\,(+\gamma)$
(*f*) ${}^{12}_{6}\mathrm{C} + {}^{2}_{1}\mathrm{D} = {}^{13}_{7}\mathrm{N} + {}^{1}_{0}n$
(*g*) ${}^{23}_{11}\mathrm{Na} + {}^{2}_{1}\mathrm{D} = {}^{24}_{11}\mathrm{Na} + {}^{1}_{1}\mathrm{H}$
(*h*) ${}^{27}_{13}\mathrm{Al} + {}^{2}_{1}\mathrm{D} = {}^{25}_{12}\mathrm{Mg} + {}^{4}_{2}\mathrm{He}$
(*i*) ${}^{27}_{13}\mathrm{Al} + {}^{1}_{0}n = {}^{24}_{11}\mathrm{Na} + {}^{4}_{2}\mathrm{He}$
(*j*) ${}^{27}_{13}\mathrm{Al} + {}^{1}_{0}n = {}^{27}_{12}\mathrm{Mg} + {}^{1}_{1}\mathrm{H}$
(*k*) ${}^{27}_{13}\mathrm{Al} + {}^{1}_{0}n = {}^{28}_{13}\mathrm{Al}\,(+\gamma)$
(*l*) ${}^{9}_{4}\mathrm{Be} + \gamma = {}^{8}_{4}\mathrm{Be} + {}^{1}_{0}n$
(*m*) ${}^{7}_{3}\mathrm{Li} + {}^{2}_{1}\mathrm{D} = 2\,{}^{4}_{2}\mathrm{He} + {}^{1}_{0}n$
(*n*) ${}^{11}_{5}\mathrm{B} + {}^{1}_{1}\mathrm{H} = 3\,{}^{4}_{2}\mathrm{He}$
(*o*) ${}^{10}_{5}\mathrm{B} + {}^{2}_{1}\mathrm{D} = 3\,{}^{4}_{2}\mathrm{He}$
(*p*) ${}^{44}_{20}\mathrm{Ca} + {}^{1}_{1}\mathrm{H} = {}^{44}_{21}\mathrm{Sc} + {}^{1}_{0}n$
(*q*) ${}^{63}_{29}\mathrm{Cu} + {}^{1}_{0}n = {}^{62}_{29}\mathrm{Cu} + 2\,{}^{1}_{0}n$

The possibility of a transmutation of the nucleus as a result of bombardment by particles rich in energy was first established in the case designated (*a*) above [1].

1. Nuclear Disruption caused by α-Rays

Magnesium bombarded by the α-rays from ThC′ emits three groups of protons [2] having the ranges 31, 40·5, and 52 cm. All three groups of protons are emitted in the capture of α-particles by the most abundant isotope of magnesium, ${}^{24}\mathrm{Mg}$, and ${}^{27}\mathrm{Al}$ is produced. The difference between the mass defect of ${}^{24}\mathrm{Mg}$ *plus* the mass and energy of the α-particle and the mass defect of ${}^{27}\mathrm{Al}$ *plus* the mass and energy of the proton emitted gives the amount of energy absorbed in the transmutation. The protons of range 52 cm. are emitted when the resulting ${}^{27}\mathrm{Al}$ nucleus is in the ground state, whereas for the other two ranges the resulting ${}^{27}\mathrm{Al}$ nucleus is in an excited state. The corresponding amounts of energy in the latter cases which remain for the time being in the nucleus are equal to the difference in energy of protons of range 52 cm. and 40·5 cm. on the one hand, and of 52 cm. and 31 cm. on the other.

In many cases the yield of disrupted atoms increases with the energy of the disrupting particle, but this is by no means always the case. Thus for the disruption of aluminium nuclei it is found that there are certain energy values (resonance levels) of the incident α-particles which are especially effective [3, 4]. In the bombardment of aluminium with α-particles of range 3·9 cm. protons of range up to 66 cm. are emitted. If the range of the α-radiation is reduced to 3·7 cm. by the introduction of an absorbing layer, say by an atmosphere of carbon dioxide, the number of protons emitted by the aluminium nuclei shows a marked diminution. On further reducing the range to 3·45 cm. the emission of a relatively large number of protons is again observed, but now they only have a range up to 61 cm. Two additional resonance positions of this kind are found for ranges of the incident α-particles of values 3·1 and 2·7 cm. respectively. A closer investigation showed that the emitted protons in each case consist of two homogeneous groups, one of larger and one of smaller energy, as can be seen from Table 28 [3]. A proton of larger energy is emitted when the captured α-particle takes up a position in the lowest energy level in the nucleus, whereas the proton emitted has a smaller energy value when the α-particle takes up a transitory position in a higher energy level. The difference in energy is emitted in the form of γ-rays when the α-particle passes over, in the course of time, into the lowest energy level of the aluminium nucleus.

TABLE 28

Protons produced by the α-Bombardment of Aluminium

Effective α-particle		*Range of protons ejected (in cm.)*
Range (in cm.)	*Energy (in volts)*	
3·9	$5{\cdot}25\times10^6$	66 and 34
3·45	$4{\cdot}86\times10^6$	61 „ 30·5
3·1	$4{\cdot}49\times10^6$	55 „ 26·5
2·7	$4{\cdot}0\times10^6$	49 „ 22

The phenomenon described is only detected when thin aluminium foils are used. If the retardation of the α-particles takes place in a thick sheet of aluminium, all eight groups of protons appear simultaneously.

This resonance phenomenon, according to which the yield of disrupted atoms is especially large for definite energy values of the disrupting α-particles, is noticeable only with relatively slow α-particles, and this is explained somewhat in the following way. α-particles rich in energy are able to surmount the potential barrier, as in the case of α-particles of energy 3·5 million volts when they are incident on beryllium nuclei. Slower α-particles, on the other hand, being less energetic, are forced to find their way through the potential barrier, and they can only do this to any appreciable extent when resonance levels exist. For beryllium there are such levels at 2·5 and 1·4 million volts respectively, so that α-particles with these amounts of energy are able to traverse the barrier with relative ease.

2. Nuclear Disruption caused by Protons and by Deuterons

The behaviour of lithium and of boron has been investigated very thoroughly with respect to positive rays of hydrogen, i.e. protons of different energy [5]. Disruption of lithium under emission of α-rays has been detected even for a value of the incident energy of 20 e.kV., but with increasing energy of the H-particles (cf. Fig. 39) the amount of disruption rises rapidly. As the energy of the protons is raised from 200 to 500 e.kV. the yield of disrupted atoms shows a ninefold increase, and as the energy is raised to 1,200 e.kV. a further c. fivefold increase in the yield is noted; the value of the yield observed is here 5×10^{-8}. The highest yields are found in the bombardment of deuterium with positive rays of heavy hydrogen; for incident rays of energy 1,000 e.kV. the yield of disrupted atoms amounts to one in a few thousands, and the phenomenon of disruption is observed for a value of the energy as low as 20 e.kV.

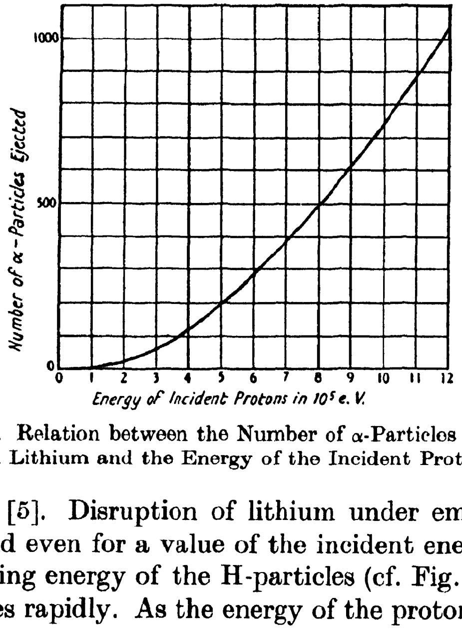

FIG. 39. Relation between the Number of α-Particles ejected from Lithium and the Energy of the Incident Protons.

The phenomenon of resonance described on p. 109 has also been observed in a few cases of disruption by accelerated protons. In the disruptive process ${}^{7}_{3}\mathrm{Li}+{}^{1}_{1}\mathrm{H} = {}^{8}_{4}\mathrm{Be}\ (+\gamma)$, when very thin films of lithium are bombarded by protons, the yield increases very rapidly with increasing energy of the protons, attains a maximum at 0·45 e.kV., and then falls rapidly as the energy is still further increased.

The majority of the α-particles ejected when lithium is bombarded with protons have a range of 8·4 cm., but particles are also present which have shorter ranges (0·5 to 1·7 cm.) and a similar frequency of occurrence. These latter particles arise from the process ${}^{6}_{3}\mathrm{Li}+{}^{1}_{1}\mathrm{H} = {}^{4}_{2}\mathrm{He}+{}^{3}_{2}\mathrm{He}$, whereas the former are due to the disruption of ${}^{7}_{3}\mathrm{Li}$. In the disruption of lithium by deuterons, α-particles of range 13·2 cm. are observed, as well as a group about ten times as strong with a continuous distribution of range between 1 and 7·8 cm.; the former are the result of the process ${}^{6}_{3}\mathrm{Li}+{}^{2}_{1}\mathrm{D} = 2\,{}^{4}_{2}\mathrm{He}$, whereas the latter are due to the process ${}^{7}_{3}\mathrm{Li}+{}^{2}_{1}\mathrm{D} = 2\,{}^{4}_{2}\mathrm{He}+{}^{1}_{0}n$. In addition, when the disruption takes place in accordance with the scheme ${}^{6}_{3}\mathrm{Li}+{}^{2}_{1}\mathrm{D} = {}^{7}_{3}\mathrm{Li}+{}^{1}_{1}\mathrm{H}$, protons are also observed with a range of 30·5 cm. for deuterons of energy 500 e.kV. The emission of α-rays of range 8·4 cm. by lithium when it is bombarded by protonic rays takes place in accordance with the expression

$${}^{7}_{3}\mathrm{Li}+{}^{1}_{1}\mathrm{H} = {}^{8}_{4}\mathrm{Be} = 2\,{}^{4}_{2}\mathrm{He},\dagger$$

or $$7{\cdot}0180+1{\cdot}0081 = 8{\cdot}0078+0{\cdot}0183 = 2\times 4{\cdot}0039+0{\cdot}0183.$$

We thus see that a loss of mass of 0·0183 takes place in the process.

Now the energy‡ corresponding to a mass-difference of 0·0183 units of atomic weight has the value $17{\cdot}04\times 10^{6}$ e.V., so that we should expect this amount of energy to be released in the above process. Just this amount of energy, more accurately $16{\cdot}97\times 10^{6}$ e.V., is found in the form of the kinetic energy of the He-ions (α-particles) produced.

Interesting cases of atomic disruption are presented by the effect of deutronic rays on heavy hydrogen [6] (deuterium); these are as follows:

$${}^{2}_{1}\mathrm{D}+{}^{2}_{1}\mathrm{D} = {}^{3}_{1}\mathrm{T}+{}^{1}_{1}\mathrm{H}, \qquad {}^{2}_{1}\mathrm{D}+{}^{2}_{1}\mathrm{D} = {}^{3}_{2}\mathrm{He}+{}^{1}_{0}n.$$

We thus see that these processes lead to the formation of isotopes of hydrogen of mass 1 and 3, an isotope of helium of mass 3, and neutrons.

3. Nuclear Disruption caused by Neutrons

Practically all the elements can be transformed by bombardment with neutrons [7]. As the atomic number of the elements gets larger, the nuclear charge also increases, and this progressively protects the nuclei of heavy atoms from the influence of incident charged particles. For this reason elements of higher atomic number than say calcium can hardly be affected by bombardment with α-particles. Neutrons,

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† The nuclei of all the ${}^{8}_{4}\mathrm{Be}$ atoms so produced are in a highly excited state, but only about one-half of them disrupt into two α-particles; the remainder emit their excess energy in the form of γ-rays of energy 17×10^{6} e.V. (cf. p. 123).

‡ According to the theory of relativity there exists between the energy content (E) and the mass (M) of a system the simple relation $M = E/c^{2}$, where c represents the velocity of light. (See also Table 57, p. 294.)

on the other hand, having no charge, are able to reach even the heaviest nuclei and to become incorporated in them. In a consideration of atomic transmutation under the action of neutrons it is necessary to distinguish between three cases:

(*a*) The admission of the neutron into the nucleus is accompanied by the ejection of a proton, so that the atomic number is reduced by one unit, whilst the nuclear mass remains unchanged.

(*b*) The admission of the neutron into the nucleus is accompanied by the ejection of an α-particle, so that the resulting element has an atomic number smaller by two units than the original element, and a nuclear mass smaller by three units.

(*c*) The admission of the neutron is not accompanied by the ejection of a nuclear constituent. The neutron is simply captured by the nucleus, and the result is the formation of a higher isotope of the element under consideration.

These three possibilities are well illustrated by considering the behaviour of the element aluminium:

$$(a)\ {}^{27}_{13}\mathrm{Al}+{}^{1}_{0}n \longrightarrow {}^{27}_{12}\mathrm{Mg}+{}^{1}_{1}\mathrm{H}, \qquad (b)\ {}^{27}_{13}\mathrm{Al}+{}^{1}_{0}n \longrightarrow {}^{24}_{11}\mathrm{Na}+{}^{4}_{2}\mathrm{He},$$

$$(c)\ {}^{27}_{13}\mathrm{Al}+{}^{1}_{0}n \longrightarrow {}^{28}_{13}\mathrm{Al}.$$

To bring about the processes designated (*a*) and (*b*) it is necessary to use neutrons of high energy value, for the reaction (*a*) can only take place when the sum of the masses of ${}^{27}_{13}\mathrm{Al}$ and ${}^{1}_{0}n$ *plus* the equivalent mass of the energy of the neutron exceeds the sum of the masses of ${}^{27}_{12}\mathrm{Mg}$ and the proton *plus* the equivalent mass of the kinetic energy of the proton. For this to be fulfilled it is necessary that the kinetic energy of the neutron shall have a value of several million electron volts. The third case (*c*) is different. This process is exothermic and can take place even in the entire absence of kinetic energy. Since slow neutrons are captured much more readily than fast ones, the process (*c*) proceeds by preference under the influence of slow neutrons. It has been seen on p. 76 that these slow neutrons are produced by allowing fast neutrons, such as those emitted by a radon-beryllium source, to pass through water, paraffin wax, or other substances containing hydrogen. The process (*c*) is sometimes designated 'hydrogen sensitive' and can be encouraged by surrounding the neutron source for this purpose with a layer of water or paraffin wax several centimetres in thickness.

The process of nuclear disruption is usually studied by making use of a Wilson cloud chamber, or of the effect in an ionization chamber or a counter device of the ionizing particles produced during the disruption process. In the case of the reaction

$${}^{10}_{5}\mathrm{B}+{}^{1}_{0}n = {}^{11}_{5}\mathrm{B} = {}^{7}_{3}\mathrm{Li}+{}^{4}_{2}\mathrm{He}$$

it has been possible to establish the formation of helium by optical spectroscopy. From the amount of helium determined in this work it was calculated that a beryllium-radon source containing 1 milli-curie of radon emits at least $6{\cdot}7\times10^3$ neutrons per second [8].

In many cases the products of transformation effected by neutrons are not new stable atoms but unstable radioactive atoms. Such radioactive substances have been produced in large numbers by neutron bombardment.

The first method used for the production of artificial radioactive elements was the bombardment with α-particles. This was soon replaced by the method of neutron bombardment, which has a much larger field of application, and the technique of which is much easier. More recently, however, a very potent third method (the cyclotron, p. 290) has been introduced. In this method the elements are bombarded by deuterons, and it is so effective that it is not unlikely that in the future it will largely supersede all other methods. (See p. 114 et seq.)

4. Nuclear Disruption caused by γ-Rays

In a few cases it has been possible to bring about an alteration of the nucleus by the action of γ-rays from the radium and thorium series [9]. Thus deuterium, which is a relatively weak structure, can be destroyed by the action of γ-rays, whereby an atom of heavy hydrogen is converted into a light hydrogen atom and a neutron, in accordance with the following expression:

$$^{2}_{1}\mathrm{D}+\gamma = {}^{1}_{1}\mathrm{H}+{}^{1}_{0}n.$$

If the γ-rays from ThC″ are used, which possess an energy quantum of value $2{\cdot}65\times10^6$ e.V., particles $^{1}_{1}\mathrm{H}$ and $^{1}_{0}n$ are obtained, each of energy $0{\cdot}25\times10^6$ e.V. From this it follows that the destruction of the deuterium nucleus requires an amount of energy given by $2{\cdot}1\times10^6$ e.V. The element $^{9}_{4}\mathrm{Be}$ also can be disrupted by the γ-rays from radium or thorium products, in accordance with the scheme:

$$^{9}_{4}\mathrm{Be}+\gamma = {}^{8}_{4}\mathrm{Be}+{}^{1}_{0}n.$$

In this process an amount of energy of $1{\cdot}6\times10^6$ e.V. suffices for the removal of a neutron from $^{9}_{4}\mathrm{Be}$. It should be mentioned that the above equation representing the action of γ-rays on heavy hydrogen is reversible; it has been possible to observe the union of neutrons with protons to the accompaniment of γ-ray emission. The nuclear disruption caused by very hard γ-rays produced artificially is discussed on p. 123.

ARTIFICIAL RADIOACTIVITY

5. Production of Artificial Radioactivity by Neutrons

Many kinds of atoms arise from the action of neutrons on other atomic types, and quite a number of them are unstable. These generally emit β-rays when they disintegrate, and the resulting products are stable atoms of another element. In order to illustrate the processes of disintegration of atoms rendered radioactive by artificial means we shall consider the case of the activation of aluminium discussed on p. 112. In this process the following three isotopes of elements are produced from aluminium when it is bombarded by neutrons: ${}^{27}_{12}\mathrm{Mg}$, ${}^{24}_{11}\mathrm{Na}$, ${}^{28}_{13}\mathrm{Al}$. All three of these products are radioactive, and they disintegrate in accordance with the following schemes:

$$
\begin{aligned}
{}^{27}_{12}\mathrm{Mg} &= {}^{27}_{13}\mathrm{Al} + \beta \quad (T = 12 \text{ min.}),\\
{}^{24}_{11}\mathrm{Na} &= {}^{24}_{12}\mathrm{Mg} + \beta \quad (T = 15 \text{ hours}),\\
{}^{28}_{13}\mathrm{Al} &= {}^{28}_{14}\mathrm{Si} + \beta \quad (T = 2{\cdot}3 \text{ min.}).
\end{aligned}
$$

Of these three processes the first is the most interesting, in so far as it leads to the formation of the only stable isotope of aluminium ${}^{27}_{13}\mathrm{Al}$, the element originally bombarded by neutrons. By means of neutrons it is possible in this case temporarily to suppress the existence of the stable isotope of aluminium, but this initial element is formed again, the half-value period of growth being 12 minutes. On the other hand, in the other two cases of the transmutation of aluminium, known isotopes of Mg and Si are the resulting end-products.

Experiment shows that the same mass-number (atomic mass) is not associated with two atomic types of the same element, one of them being stable and the other active or unstable. Thus ${}^{53}_{24}\mathrm{Cr}$ is a stable type of chromium. The union of a neutron with the most abundant isotope of chromium ${}^{52}_{24}\mathrm{Cr}$ does not result in the formation of a radioactive isotope, but of the stable isotope ${}^{53}_{24}\mathrm{Cr}$, for the existence of the latter excludes that of a radioactive isotope ${}^{53}_{24}\mathrm{Cr}$. Most of the elements of even atomic number have numerous isotopes, and the union of neutrons with such elements leads in many cases of necessity to stable higher isotopes of the same element having a higher atomic mass. The occurrence of such processes can be established by absorption experiments (cf. p. 112). In all cases where absorption of neutrons is observed and artificial radioactivity does not result from the process, it is possible to conclude that a stable isotope or an isotope of long life has been formed by the act of absorption. Moreover, such a process is often associated with the emission of γ-rays in detectable amount, for the energy liberated by the addition of neutrons is emitted in this form.

When neutrons are captured and this is accompanied by the emission of α-rays, the result is an element of lower atomic number. The latter processes, as well as those in which protons are emitted, have only been observed for the lighter elements up to and including zinc, when neutrons emitted by radium-beryllium (or radon-beryllium) sources have been used. Under the action of such particles, the majority of the elements are found only to form a simple union with neutrons. By means of bombardment with deuterons and other particles of very large energy, it has been possible to observe the emission of material particles after the capture of neutrons even by the heavier elements.

The radioactive isotopes produced artificially by neutrons are shown in Table 29, so far as they are known, and their half-value periods are also included in the table. It begins with the element lithium, since for elements with an atomic number less than 3 no activity has been found as a result of their bombardment with neutrons. The chief isotopes of the elements bombarded by neutrons are printed in italics.

An examination of the table reveals that the observed periods lie between a few seconds and a few years. Energy considerations render it improbable that numerous undiscovered radioactive types of atoms exist, which decay with a period of only a fraction of a second. On the other hand, numerous long-lived radioactive isotopes may be capable of existence, their periods of life being too short for them to be still available should they have been formed at a time when the substances under consideration were subjected to bombardment by neutrons in another part of the universe, and too long for them to be produced in measurable quantity by the action of radiation in the laboratory during the course of days or weeks. The element ${}^{40}_{19}\mathrm{K}$, which is responsible for the natural radioactivity of potassium, has a period of life sufficiently large to ensure that it has not yet completely disintegrated in terrestrial material in the course of geological time [10]. Here we have a radioactive isotope of very long life which was produced in pre-terrestrial times by means of the process

$${}^{39}_{19}\mathrm{K}+{}^{1}_{0}n = {}^{40}_{19}\mathrm{K},$$

or by virtue of an analogous reaction such as

$${}^{40}_{20}\mathrm{Ca}+{}^{1}_{0}n = {}^{40}_{19}\mathrm{K}+{}^{1}_{1}\mathrm{H}.$$

✓That one and the same radioactive isotope can be produced in different ways is likewise clearly revealed in Table 29. Thus the isotope ${}^{28}_{13}\mathrm{Al}$ can be prepared either from Al, Si, or P, and the isotope ${}^{52}_{23}\mathrm{V}$ from V, Cr, or Mn. The result of the attachment of neutrons to the nuclei of thorium [12] and uranium [7, 13] calls for special mention. In the first case a new

TABLE 29. (Cf. Table 56, p. 292)

Radioactive Isotopes produced by Neutron Bombardment of the Elements [11]

Element bombarded	*Mass-numbers of stable isotopes of element bombarded*	*Resulting radioactive isotope*	*Half-value period of resulting isotope*
3 Li	6, *7*	${}^{8}_{3}\mathrm{Li}$	0·8 s.
4 Be	8, *9*, 10	${}^{6}_{2}\mathrm{He}$	1 s.
8 O	*16*, 17, 18	${}^{16}_{7}\mathrm{N}$	9 s.
9 F	*19*	${}^{16}_{7}\mathrm{N}$; ${}^{19}_{8}\mathrm{O}$; ${}^{20}_{9}\mathrm{F}$	9 s.; 31 s.; 8 s.
10 Ne	*20*, 21, 22		
11 Na	*23*	${}^{23}_{10}\mathrm{Ne}$; ${}^{24}_{11}\mathrm{Na}$	33 s.; 15 h.
12 Mg	*24*, 25, 26	${}^{23}_{10}\mathrm{Ne}$; ${}^{27}_{12}\mathrm{Mg}$; ${}^{24}_{11}\mathrm{Na}$	33 s.; 12 m.; 15 h.
13 Al	*27*	${}^{28}_{13}\mathrm{Al}$; ${}^{27}_{12}\mathrm{Mg}$; ${}^{24}_{11}\mathrm{Na}$	2·3 m.; 12 m.; 15 h.
14 Si	*28*, 29, 30	${}^{28}_{13}\mathrm{Al}$; ${}^{31}_{14}\mathrm{Si}$	2·3 m.; 2·5 h.
15 P	*31*	${}^{28}_{13}\mathrm{Al}$; ${}^{31}_{14}\mathrm{Si}$; ${}^{32}_{15}\mathrm{P}$	2·3 m.; 2·5 h.; 14·5 d.
16 S	*32*, 33, 34	${}^{32}_{15}\mathrm{P}$	14·5 d.
17 Cl	*35*, *37*	${}^{38}_{17}\mathrm{Cl}$†; ${}^{32}_{15}\mathrm{P}$†; ${}^{35}_{16}\mathrm{S}$	38 m.; 14·5 d.; 80 d.
18 A	36, 38, *40*	${}^{41}_{18}\mathrm{A}$	110 m.
19 K	*39*, 40, 41	${}^{42}_{19}\mathrm{K}$; ${}^{41}_{18}\mathrm{A}$	12·5 h.; 110 m.
20 Ca	*40*, 42, 43, 44	${}^{45}_{20}\mathrm{Ca}$†; ${}^{42}_{19}\mathrm{K}$	2·3 h.; 12·5 h.
21 Sc	*45*	${}^{42}_{19}\mathrm{K}$; ${}^{46}_{21}\mathrm{Sc}$	12·5 h.; 90 d.
22 Ti	46, 47, *48*, 49, 50	${}^{51}_{22}\mathrm{Ti}$; ${}^{45}_{20}\mathrm{Ca}$	3 m.; 2·3 h.
23 V	*51*	${}^{52}_{23}\mathrm{V}$	3·8 m.
24 Cr	50, *52*, 53, 54	${}^{52}_{23}\mathrm{V}$	3·8 m.
25 Mn	*55*	${}^{52}_{23}\mathrm{V}$; ${}^{56}_{25}\mathrm{Mn}$	3·8 m.; 2·5 h.
26 Fe	54, *56*, 57	${}^{56}_{25}\mathrm{Mn}$	2·5 h.
27 Co	57, *59*	${}^{60}_{27}\mathrm{Co}$; ${}^{56}_{25}\mathrm{Mn}$; ${}^{59}_{26}\mathrm{Fe}$	*c.* 1 y.; 2·5 h.; 40 d.
28 Ni	*58*, *60*, 61, 62, 64	${}^{58}_{27}\mathrm{Co}$; ${}^{60}_{27}\mathrm{Co}$; ${}^{63}_{28}\mathrm{Ni}$	11 m.; *c.* 1 y.; 160 m.
29 Cu	*63*, *65*	${}^{66}_{29}\mathrm{Cu}$; ${}^{64}_{29}\mathrm{Cu}$†; ${}^{63}_{28}\mathrm{Ni}$	5 m.; 12·5 h.; 160 m.
30 Zn	*64*, *66*, 67, *68*, 70	${}^{66}_{29}\mathrm{Cu}$; ${}^{64}_{29}\mathrm{Cu}$†	5 m.; 12·5 h.
31 Ga	*69*, *71*	${}^{70}_{31}\mathrm{Ga}$; ${}^{72}_{31}\mathrm{Ga}$	20 m.; 23 h.
32 Ge	*70*, *72*, 73, *74*, 76	${}^{75}_{32}\mathrm{Ge}$†; ${}^{?}_{32}\mathrm{Ge}$†	20 h.; 30 m.
33 As	*75*	${}^{76}_{33}\mathrm{As}$	26 h.
34 Se	74, 76, 77, *78*, *80*, 82	${}^{?}_{34}\mathrm{Se}$	1 h.
35 Br	*79*, *81*	${}^{80}_{35}\mathrm{Br}$; ${}^{80}_{35}\mathrm{Br}$; ${}^{82}_{35}\mathrm{Br}$	18 m.; 4·5 h.; 34 h.
36 Kr	78, 80, 82, 83, *84*, 86	?; ?; ?	18 h.; 4·5 h.; 74 m.
37 Rb	*85*, *87*	${}^{86}_{37}\mathrm{Rb}$†; ${}^{88}_{37}\mathrm{Rb}$†	18 m.; 18 d.
38 Sr	84, 86, 87, *88*	${}^{89}_{38}\mathrm{Sr}$	3 h. and 55 d.
39 Y	*89*	${}^{90}_{39}\mathrm{Y}$	70 h.
40 Zr	*90*, 91, *92*, *94*, 96	${}^{97}_{40}\mathrm{Zr}$	44 h.
41 Nb	*93*		
42 Mo	92, 94, *95*, *96*, 97, *98*, 100	${}^{?}_{42}\mathrm{Mo}$; ${}^{?}_{42}\mathrm{Mo}$; ${}^{?}_{42}\mathrm{Mo}$	2·5 m.; 17 m.; 36 h.
44 Ru	96, 99, 100, *101*, *102*, 104	${}^{?}_{44}\mathrm{Ru}$; ${}^{?}_{44}\mathrm{Ru}$; ${}^{?}_{44}\mathrm{Ru}$	100 s.; 11 h.; 170 h.
45 Rh	101, *103*	${}^{?}_{45}\mathrm{Rh}$; ${}^{?}_{45}\mathrm{Rh}$	44 s.; 3·9 m.
46 Pd	102, 104, *105*, *106*, *108*, 110	${}^{?}_{46}\mathrm{Pd}$; ${}^{?}_{46}\mathrm{Pd}$; ${}^{?}_{46}\mathrm{Pd}$	3 m.; 12 h.; 60 h.
47 Ag	*107*, *109*	${}^{106}_{47}\mathrm{Ag}$; ${}^{110}_{47}\mathrm{Ag}$†; ${}^{108}_{47}\mathrm{Ag}$†	24·5 m. and 8·2 d.; 22 s.; 2·3 m.
48 Cd	106, 108, *110*, *111*, *112*, *113*, 116	${}^{?}_{48}\mathrm{Cd}$; ${}^{?}_{48}\mathrm{Cd}$; ${}^{?}_{48}\mathrm{Cd}$; ${}^{106}_{47}\mathrm{Ag}$; ${}^{108}_{47}\mathrm{Ag}$; ${}^{110}_{47}\mathrm{Ag}$; ${}^{111}_{47}\mathrm{Ag}$; ${}^{112}_{47}\mathrm{Ag}$	33 m.; 4·3 h.; 58 h.; 24·5 m. and 8·2 d.; 2·3 m.; 22 s.; 7·5 d.; 3·2 h.
49 In	113, *115*	${}^{116}_{49}\mathrm{In}$; ${}^{116}_{49}\mathrm{In}$; ${}^{112}_{47}\mathrm{Ag}$	13 s.; 54 m.; 3.2 h.

TABLE 29 (*cont.*)

Element bombarded	*Mass-numbers of stable isotopes of element bombarded*	*Resulting radioactive isotope*	*Half-value period of resulting isotope*
50 Sn	112, 114, 115, 116, 117, *118*, 119, *120*, 122, 124	${}^{?}_{50}\mathrm{Sn}$; ${}^{?}_{50}\mathrm{Sn}$	8 m.; 18 m.
51 Sb	*121*, *123*	${}^{120}_{51}\mathrm{Sb}$; ${}^{122}_{51}\mathrm{Sb}$; ${}^{124}_{51}\mathrm{Sb}$	16 m.; 2·5 d.; 60 d.
52 Te	122, 123, 124, 125, *126*, *128*, *130*	${}^{?}_{52}\mathrm{Te}$	1·1 h.
53 I	*127*	${}^{128}_{53}\mathrm{I}$	25 m.
55 Cs	*133*	${}^{134}_{55}\mathrm{Cs}$	1·5 h.
56 Ba	130, 132, 134, 135, 136, 137, *138*	—; ${}^{139}_{56}\mathrm{Ba}$†	2·5 m.; 80 m.
57 La	*139*	${}^{140}_{57}\mathrm{La}$	1·3 d.
58 Ce	*140*, 142		
59 Pr	*141*	${}^{140}_{59}\mathrm{Pr}$; ${}^{142}_{59}\mathrm{Pr}$	3 m.; 19 h.
60 Nd	*142*, 143, *144*, 145, 146 148, 150	${}^{147}_{60}\mathrm{Nd}$†	2 h.
62 Sm	144, 147, 148, 149, 150 *152*, *154*	${}^{151}_{62}\mathrm{Sm}$; ${}^{153}_{62}\mathrm{Sm}$	21 m.; 2 d.
63 Eu	*151*, *153*	${}^{154}_{63}\mathrm{Eu}$†	9·3 h.
64 Gd	*155*, *156*, 157, *158*, 160		
65 Tb	*159*	${}^{160}_{65}\mathrm{Tb}$	3·9 h.
66 Dy	*161*, *162*, *163*, *164*	${}^{165}_{66}\mathrm{Dy}$	2·5 h.
67 Ho	*165*	${}^{166}_{67}\mathrm{Ho}$	30 h.
68 Er	*166*, *167*, *168*, 170	${}^{171}_{68}\mathrm{Er}$†; ${}^{169}_{68}\mathrm{Er}$†	7 m.; 12 h.
69 Tm	*169*	${}^{170}_{69}\mathrm{Tm}$	120 d.
70 Yb	171, *172*, 173, *174*, 176	${}^{175}_{70}\mathrm{Yb}$†	2·1 h.
71 Lu	*175*, 177	${}^{176}_{71}\mathrm{Lu}$; ${}^{178}_{71}\mathrm{Lu}$†	4 h.; 8 d.
72 Hf	176, 177, *178*, 179, *180*	${}^{181}_{72}\mathrm{Hf}$	55 d.
73 Ta	*181*	${}^{180}_{73}\mathrm{Ta}$‡; ${}^{182}_{73}\mathrm{Ta}$	8 h.; 97 d.
74 W	*182*, 183, *184*, *186*	?	1 d.
75 Re	*185*, *187*	${}^{?}_{75}\mathrm{Re}$; ${}^{?}_{75}\mathrm{Re}$; ${}^{?}_{75}\mathrm{Re}$	20 m.; 18 h.; 85 h.
76 Os	186, 187, 188, 189, *190* *192*	${}^{?}_{76}\mathrm{Os}$	40 h.
77 Ir	191, *193*	${}^{192}_{77}\mathrm{Ir}$; ${}^{194}_{77}\mathrm{Ir}$; ${}^{?}_{77}\mathrm{Ir}$; ${}^{?}_{77}\mathrm{Ir}$	2 m.; 1·5 m. and 19 h.; 28 m.; 8·5 h.
78 Pt	192, 194, *195*, *196*, 198	${}^{197}_{78}\mathrm{Pt}$; ${}^{199}_{78}\mathrm{Pt}$	18 h. and 3·3 d.; 50 m.
79 Au	*197*	${}^{198}_{79}\mathrm{Au}$	2·7 d.
80 Hg	196, 197, 198, 199, *200*, 201, *202*, 203, 204	${}^{205}_{80}\mathrm{Hg}$	41 h.
81 Tl	203, *205*	${}^{204}_{81}\mathrm{Tl}$†; ${}^{206}_{81}\mathrm{Tl}$†	5 m.; 97 m.
82 Pb	204, *206*, *207*, *208*	${}^{209}_{82}\mathrm{Pb}$	3 h.
90 Th	*232*	${}^{229}_{88}\mathrm{Ra}$; ${}^{233}_{91}\mathrm{Pa}$; ${}^{231}_{89}\mathrm{Ac}$; ${}^{233}_{90}\mathrm{Th}$; ${}^{233}_{89}\mathrm{Ac}$	1 m.; 2·5 m.; 12 m.; 28 m.; 3·5 h.
92 U	235, *238*	${}^{236}_{92}\mathrm{U}$; ${}^{239}_{92}\mathrm{U}$	23 m.; 10 s. and 40 s. (see below)

† Indicates uncertainty; thus the resulting isotope may have another mass; or in reality ${}^{36}\mathrm{Cl}$ may be produced instead of ${}^{38}\mathrm{Cl}$, and so on.

‡ Emits 2 neutrons after the capture of 1 fast neutron.

radioactive transformation series is produced (see Table 29), and hence new isotopes of Pa, Ac, Ra, and so on, whereas in the case of uranium, elements of atomic number higher than that of uranium 92 are formed, some of which are higher homologues of rhenium, osmium, iridium, and platinum. The following are the reactions observed:

$$(a)\quad {}_{92}\mathrm{U}+{}_{0}n\begin{pmatrix}\text{slow}\\ \text{or}\\ \text{fast}\end{pmatrix}\longrightarrow({}_{92}\mathrm{U}+n)\xrightarrow[(10\text{s.})]{\beta}{}_{93}\text{Eka-Re}\xrightarrow[(2{\cdot}2\text{m.})]{\beta}{}_{94}\text{Eka-Os}\longrightarrow$$

$$\xrightarrow[(59\text{m.})]{\beta}{}_{95}\text{Eka-Ir}\xrightarrow[(66\text{h.})]{\beta}{}_{96}\text{Eka-Pt}\xrightarrow[(2{\cdot}5\text{h.})]{\beta}{}_{97}\text{Eka-Au(?)}.$$

$$(b)\quad {}_{92}\mathrm{U}+{}_{0}n\,(\text{fast})\longrightarrow({}_{92}\mathrm{U}+n)\xrightarrow[(40\text{s.})]{\beta}{}_{93}\text{Eka-Re}\xrightarrow[(16\text{m.})]{\beta}{}_{94}\text{Eka-Os}\longrightarrow$$

$$\xrightarrow[(5{\cdot}7\text{h.})]{\beta}{}_{95}\text{Eka-Ir}.$$

$$(c)\quad {}_{92}\mathrm{U}+{}_{0}n\,(\text{slow})\longrightarrow({}_{92}\mathrm{U}+n)\xrightarrow[(23\text{m.})]{\beta}{}_{93}\text{Eka-Re(?)}.$$

(The half-value periods of the relevant disintegrations are given in brackets beneath the arrows.)

Whereas it is necessary to use neutrons of high energy in order to produce both ${}^{27}_{12}\mathrm{Mg}$ and ${}^{24}_{11}\mathrm{Na}$ from ${}^{27}_{13}\mathrm{Al}$, this is not the case in the production of radio-aluminium ${}^{28}_{13}\mathrm{Al}$, for the last-named process gives rise to a greater yield with slow neutrons than with fast neutrons. This statement holds quite generally for the production of higher isotopes of the element bombarded, in contrast to the production of isotopes of other elements. In the former process all that happens is the incorporation of a neutron into the nucleus without ejection of an α-particle or a proton, and since such a process is an exothermic one, it does not involve the addition of energy in the form of kinetic energy of the neutrons. But when radio-elements are produced with emission of an α- or an H-particle, this is only possible by the addition of an amount of energy represented by several millions of volts in the form of kinetic energy of the bombarding neutrons. For this reason, in the preparation of higher isotopes by bombardment with neutrons, the velocity of the neutrons is very greatly reduced by the introduction of paraffin wax, water, or similar substances (cf. p. 76). The neutrons gradually give up their energy by repeated collision with the hydrogen nuclei, and the probability of capture of these quite slow neutrons is thus in many cases markedly greater than that of fast neutrons, such as are emitted by a beryllium-radium source. In this way, for instance, it is possible to attain a 40-fold yield in the preparation of radio-silver, and even a 100-fold yield in the preparation of radio-dysprosium.

From amongst the various methods that lead to the production of artificial radioactivity, that of the bombardment by neutrons is the one which results in the best yield (see, however, p. 113). All the neutrons are ultimately captured by nuclei, and by suitably devised arrangements it is possible to ensure that practically all of them are absorbed in the element whose transformation is desired. On the other hand, only a small fraction of incident α-particles or accelerated protons (cf. p. 112) reaches the nucleus, whereas the large majority of them lose their energy by interaction with the shell of the atom and can be disregarded as components of nuclear constitution. The transmutation process effected by accelerated deuterons results in individual cases in a much improved yield; here the yield can amount to as much as 1 transmuted nucleus for a few thousand deuterons.

6. Production of Artificial Radioactivity by α-Rays [14]

Most of the elements situated between boron and calcium have been activated under the influence of α-rays. After the capture of the α-particle by the atomic nucleus, this emits either a neutron or a proton and a radioactive isotope of a new element is formed. The bombardment of Mg by α-particles leads either to the formation of a radioactive $^{27}_{14}\mathrm{Si}$ in accordance with the equation

$$^{24}_{12}\mathrm{Mg}+^{4}_{2}\alpha = ^{27}_{14}\mathrm{Si}+^{1}_{0}n,$$

or of the radioactive isotope $^{28}_{13}\mathrm{Al}$ according to the equation

$$^{25}_{12}\mathrm{Mg}+^{4}_{2}\alpha = ^{28}_{13}\mathrm{Al}+^{1}_{1}\mathrm{H}.$$

Stable isotopes of another element may of course also result from such processes. Thus when Al is bombarded by α-rays the resulting product may be either the radioactive isotope $^{30}_{15}\mathrm{P}$ with emission of a neutron, or the stable isotope $^{30}_{14}\mathrm{Si}$ in agreement with the equation

$$^{27}_{13}\mathrm{Al}+^{4}_{2}\alpha = ^{30}_{14}\mathrm{Si}+^{1}_{1}\mathrm{H}.$$

Since the same mass-number cannot be associated both with an active and with a stable isotope of the same element, a stable isotope must result from the last-named process, the isotope $^{30}_{14}\mathrm{Si}$ being stable.

Certain minerals rich in the rare earths contain appreciable amounts of uranium or thorium, as well as of light elements such as beryllium or aluminium, and in the course of geological time some of the rare earth elements contained in these minerals have undergone a slight change [15]. Gadolinium, which absorbs slow neutrons very strongly, may be mentioned in this connexion.

Whereas the active isotopes resulting from bombardment by neutrons always disintegrate with emission of β-particles whose half-value thickness lies between 0·04 and 0·25 gm./cm.^{2} aluminium, the radioactive isotopes produced by bombardment with α-particles show a different behaviour. A considerable proportion of these isotopes disintegrate with emission of positrons. By incorporating the two charges of the α-particle, the resulting isotope has too many protons in proportion to its neutron content, so that it possesses too many positive charges and gets rid of these by emitting positrons. Table 30 contains all the radioactive isotopes produced by bombardment with α-particles, and in the table it is also stated whether the radioactive isotope emits a positron or an electron.

TABLE 30

Radioactive Isotopes produced by Bombardment with α-Particles

Isotope bombarded	*Particle emitted*	*Isotope produced*	*Half-value period*
${}^{10}_{5}\mathrm{B}$	*n*	${}^{13}_{7}\mathrm{N}$ (+)	11 min.
${}^{14}_{7}\mathrm{N}$	*n*	${}^{17}_{9}\mathrm{F}$ (+)	1·2 min.
${}^{19}_{9}\mathrm{F}$	*n*	${}^{22}_{11}\mathrm{Na}$ (+)	3 years
${}^{23}_{11}\mathrm{Na}$	*n*	${}^{26}_{13}\mathrm{Al}$ (+)	7 sec.
${}^{24}_{12}\mathrm{Mg}$	*n*	${}^{27}_{14}\mathrm{Si}$ (+)	6 min.
${}^{25}_{12}\mathrm{Mg}$	*p*	${}^{28}_{13}\mathrm{Al}$ (−)	2·3 min.
${}^{26}_{12}\mathrm{Mg}$	*p*	${}^{29}_{13}\mathrm{Al}$ (−)	11 min.
${}^{27}_{13}\mathrm{Al}$	*n*	${}^{30}_{15}\mathrm{P}$ (+)	3 min.
${}^{29}_{14}\mathrm{Si}$	*p*	${}^{32}_{15}\mathrm{P}$ (−)	15 days
${}^{31}_{15}\mathrm{P}$	*n*	${}^{34}_{17}\mathrm{Cl}$ (−)	33 min.
${}^{39}_{19}\mathrm{K}$	*n*	${}^{42}_{21}\mathrm{Sc}$ (−)	3 hours
${}^{40}_{20}\mathrm{Ca}$	*p*	${}^{43}_{21}\mathrm{Sc}$ (−)	4·0 hours

n = neutron; *p* = proton; (+) indicates that the isotope produced disintegrates with emission of positrons, and (−) that it disintegrates with emission of electrons.

All the known stable as well as radioactive isotopes in the interval between hydrogen and krypton are incorporated in Fig. 40. The atomic number is plotted as abscissa, whereas the ordinates represent the difference between the mass-number and twice the atomic number. The isotopes indicated by circles in heavy line are stable, and if the concentration of the relevant isotope in the mixed element is greater than 5 per cent. it is represented by a large circle, otherwise by a small one. The circles contain the mass-number, and also the half-value period when the isotope is unstable. Circles in thin line represent active isotopes; if the upper half of the circle is in broken line, ○, the active isotope disintegrates with emission of β-rays, but if the lower half of the circle is in broken line, ○, positrons are emitted during disintegration. Active isotopes that are lighter than the stable types disintegrate with emission of positrons (see Table 56); here the structure contains too many protons in proportion to neutrons, and diminishes its excessive charge number by ejection of a positive charge. On the other hand, the isotopes that disintegrate with emission of β-rays contain too many neutrons for them to be stable, and alter their neutron:proton ratio by ejecting a negative electron, which corresponds to their taking up

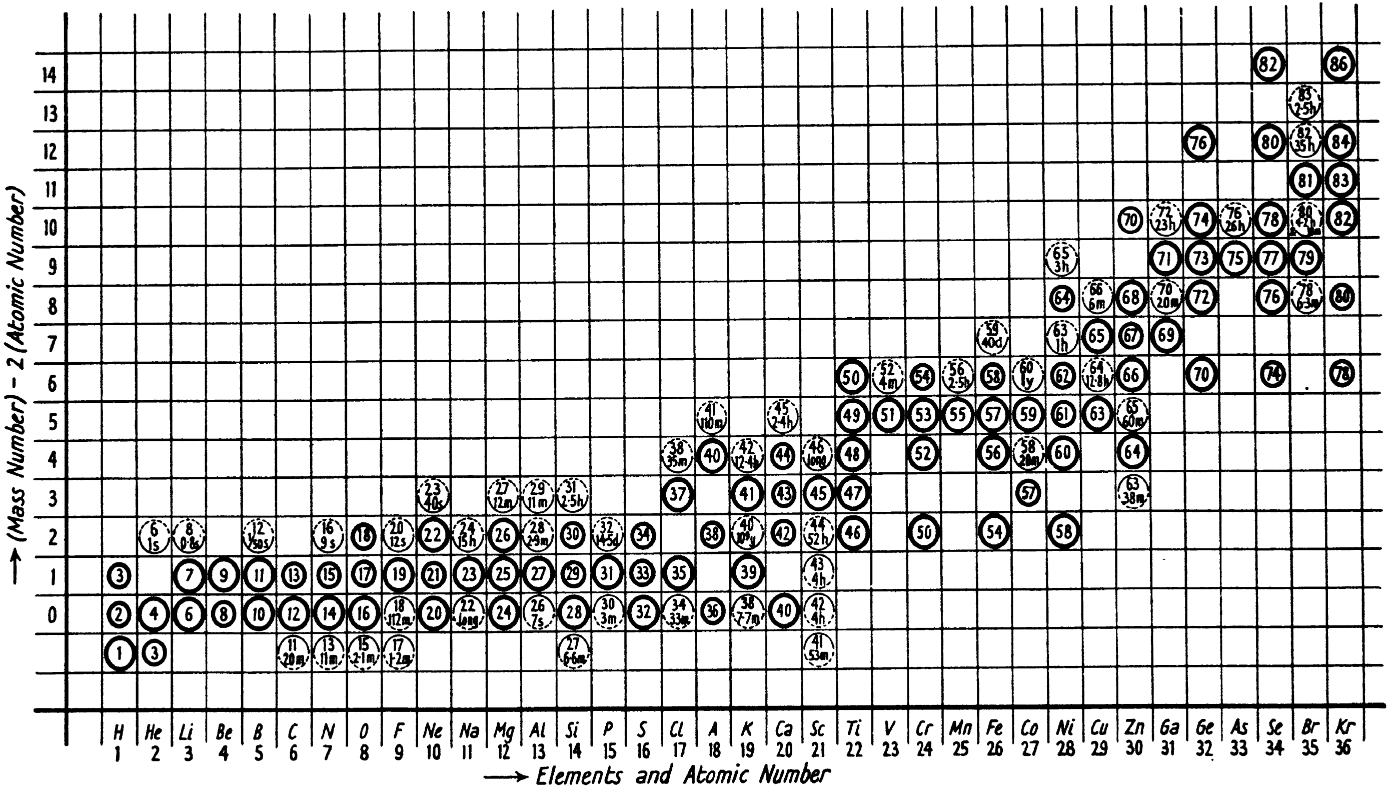

FIG. 40. Diagram incorporating most of the known Stable and Radioactive Isotopes from Hydrogen (1) to Krypton (36).
(See Tables 29 and 56.)

a positive charge. When substances are bombarded by neutrons, products are always obtained which, if unstable, contain too many neutrons, and hence disintegrate with emission of β-particles. Isotopes that disintegrate with emission of positrons have arisen as a result of the nuclei taking up α-particles or other positively charged mass-particles.

In the production of artificial radioactivity by means of α-particles the yield is a very modest one [16]. For every 10^7 α-particles of high energy value ($8{\cdot}3 \times 10^6$ e.V.) incident upon it, only 6 are effective in the activation of aluminium, i.e. in the production of radio-phosphorus ($^{30}_{15}P$), and the yield of radio-phosphorus produced falls rapidly as the range of the incident α-particles is diminished, as can be seen in Table 31.

TABLE 31

Dependence of Activation on the Energy of the Incident α-Particles

Energy of incident α-particles	*Yield*†
$8{\cdot}25 \times 10^6$ e.V.	$6{\cdot}4 \times 10^{-7}$
7·61 „	4·7 „
6·66 „	1·1 „
5·49 „	0·13 „
4·81 „	0·02 „
4·20 „	0·00 „

† Yield = probability that an α-particle of energy E will at some portion of its range form a radio-phosphorus atom.

As seen in the table, the yield in the production of radio-phosphorus is very small; that of the production of other artificial radio-elements with the aid of α-rays is still smaller.

7. Production of Artificial Radioactivity by Deuterons and γ-Rays

A number of artificial radio-elements have been generated under the influence of accelerated deuterium ions in the region between boron and aluminium [17]; representative cases are collected together in Table 32. Whereas some disintegrate with emission of positrons, others emit electrons.

When any substance is bombarded by deuterons, an activity is observed which decays with a period of 2 minutes. It is to be ascribed to the disintegration of $^{15}_{8}O$, which has been generated by the action of the deuterons on the atoms of the nitrogen $^{14}_{7}N$ in the air [18].

We have seen that neutrons are produced [9] when γ-rays are

TABLE 32

Radioactive Isotopes produced by Bombardment with Deuterons

Bombarded isotope	*Particles emitted*	*Isotope produced*	*Half-value period*
$^{10}_{5}B$	*n*	$^{11}_{6}C$ (+)	20 min.
$^{12}_{6}C$	*n*	$^{13}_{7}N$ (+)	10·5 min.
$^{14}_{7}N$	*n*	$^{15}_{8}O$ (+)	2·1 min.
$^{23}_{11}Na$	*p*	$^{24}_{11}Na$ (−)	15 hours
$^{24}_{12}Mg$	α	$^{22}_{11}Na$ (+)	3 yrs.
$^{27}_{13}Al$	*p*	$^{28}_{13}Al$ (−)	2·3 min.

allowed to impinge on beryllium or on deuterium. γ-rays are accordingly able to call forth an artificial radioactivity in the presence of these elements [19]. In consequence of the relatively small energy of the neutrons liberated, however, it is possible in this way to bring about only association processes, by the formation of a radioactive isotope of higher mass-number. In the most favourable case, when the total γ-radiation is absorbed in beryllium, the yield can attain that which would be obtained by the effect of an equivalent amount of α-rays on beryllium, i.e. by the α-rays emitted from the same quantity of radium. It should be noted, however, that in the latter case the neutrons have energies up to 10^7 e.V., and they can accordingly also produce types of atoms other than higher isotopes of the bombarded elements, as in the production by fast neutrons of $^{6}_{2}He$ from $^{9}_{4}Be$, by virtue of the reaction $^{9}_{4}Be + ^{1}_{0}n = ^{6}_{2}He + ^{4}_{2}He$.

By bombarding a lithium target with protons of minimum energy 450,000 e.V., γ-rays of very great hardness (17×10^6 e.V.) can be produced. These rays are able to bring about nuclear photo-effects by the ejection of neutrons from the nuclei of various elements such as phosphorus, copper, and bromine, the resulting radioactive isotope having a mass-number reduced by one unit [20].

It is clear from what has been said above that it is possible to produce a number of artificial radio-elements in a variety of ways. To take one example, the following methods have been successfully applied in the preparation of radio-aluminium $^{28}_{13}Al$, which decays with a half-value period of 2·3 minutes, and emits β-rays whose half-value thickness in aluminium amounts to 0·16 gm./cm.^{2}:

$$^{27}_{13}Al + ^{1}_{0}n = ^{28}_{13}Al, \qquad ^{28}_{14}Si + ^{1}_{0}n = ^{28}_{13}Al + ^{1}_{1}H,$$

$$^{31}_{15}P + ^{1}_{0}n = ^{28}_{13}Al + ^{4}_{2}He, \qquad ^{25}_{12}Mg + ^{4}_{2}\alpha = ^{28}_{13}Al + ^{1}_{1}H,$$

$$^{27}_{13}Al + ^{2}_{1}D = ^{28}_{13}Al + ^{1}_{1}H.$$

The production of the silver isotope $^{106}_{47}Ag$ can also be effected in various ways, one of which is the reaction

$$^{105}_{46}Pd + ^{2}_{1}D = ^{106}_{47}Ag + ^{1}_{0}n.$$

This isotope of silver is of interest, as it has two half-value periods, 24·5 m. and 8·2 d., the former being associated with positron emission, and the latter with the emission of β- and γ-rays, the number of γ-rays per β-ray being about 35. Here we are confronted with the phenomenon of *nuclear isomerism* [21], the branching ratio in the present instance being about 20:1 in favour of the longer period. It seems likely that the capture of orbital electrons presents an alternative to positron emission, and in the case of heavy radioactive nuclei the probability of such K-electron capture, accompanied by β- and relatively intense γ-ray emission, is theoretically about 1,000 times greater than that of the emission of positrons. The isotope of bromine $^{80}_{35}\mathrm{Br}$ also consists of radioactive isomers with half-value periods of 18 m. and 4·2 h., but in this case both isomers emit β-rays, together with γ-rays. The radioactive isotope of potassium $^{40}_{19}\mathrm{K}$, in addition to its normal β-disintegration into $^{40}_{20}\mathrm{Ca}$, probably also emits positrons with formation of $^{40}_{18}\mathrm{A}$, and some of the argon in the earth's crust and in the atmosphere may have originated in this way.

8. Accurate Determination of Atomic Weights on the Basis of Nuclear Reactions [22]

In the formulae for nuclear reactions such as

$$^{9}_{4}\mathrm{Be}+^{2}_{1}\mathrm{D} = ^{10}_{4}\mathrm{Be}+^{1}_{1}\mathrm{H}+\delta$$

we are concerned not only with the masses of the atomic types $^{9}\mathrm{Be}$, $^{2}\mathrm{D}$, $^{10}\mathrm{Be}$, and $^{1}\mathrm{H}$, but also with the mass-equivalent (δ) of the energy of the protons emitted. Moreover, a very small mass corresponds to a very large value of the energy (1 e.V. $= 1{\cdot}0741\times 10^{-9}$ units of atomic weight). In the example cited above the conversion of the energy value $\delta = 4{\cdot}749\times 10^{6}$ e.V. into units of atomic weight yields the value 0·0051, so that the measurement of the large amounts of energy that occur here corresponds to a very exact determination of the mass. It is thus possible, by the measurement of the energy of the rays emitted in nuclear reactions, as well as by the combination of various equations of nuclear reactions, to arrive at atomic weights of very great accuracy. Most of the values of the atomic weights given in Table 43, p. 182, up to oxygen, have been determined in this way (see p. 180). The numerical values are all referred to $^{16}\mathrm{O} = 16{\cdot}0000$.

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XI

APPLICATIONS OF THE THEORY OF DISINTEGRATION

1. The Fundamental Assumption

The theory of radioactive disintegration is based on the single assumption that the number of atoms of a radio-element disintegrating at any instant is always proportional to the number of atoms of that element present.

If we have 10^{11} atoms of a given radio-element and 1,000 of these disintegrate in a given interval of time, then if we have 10^{10} atoms of the same radio-element, the number disintegrating in the same time will be 100, and so on. The factor of proportionality, or the 'disintegration constant' (λ) is found to be a quantity which is entirely independent of all outside conditions such as temperature, pressure, etc., and solely characteristic of the disintegration process under consideration.

Only rarely does natural disintegration lead directly to a stable product; in most cases the disintegration of an atom is followed by a whole series of successive transformations. The conditions that arise are often very complicated, but the theory of transformation enables us to interpret them quantitatively, and to calculate the number of atoms of each intermediate product present at any time, provided that the disintegration constants are known. It also indicates means of evaluating the disintegration constants when these cannot be directly determined.

In what follows we shall deal with the application of the theory of transformation in a few typical cases of successive processes of disintegration.

2. Production of a Substance of Short Life from a Parent Substance of Long Life. Separation and Decay of UX

If we investigate the behaviour of uranium nitrate or of any other uranium salt, we find that it emits β-rays in addition to α-rays. We may recall that the radioactivity is a property of the uranium atom, and that it is entirely independent of the state of chemical combination of the element. If we submit the uranium salt to a process of crystallization or to processes of chemical separation, we find that the strength of the α-radiation from the substance is unaltered, but that the β-radiation has been influenced. It is possible, in fact, e.g. by the addition of an iron salt to the solution of the uranium compound and later precipitation of the iron by means of ammonium carbonate, to separate completely the β-radiation from the uranium. But this is now emitted with the same intensity from the precipitated iron salt. From this we may conclude that the β-radiation has its origin not in the uranium but in a radioactive product separable from uranium, and known as uranium X.

The β-radiation of the iron salt does not remain constant, but decreases exponentially; viz. after the lapse of about 24 days it has already sunk to one-half of its original intensity, after 48 days to one-quarter, in 72 days to one-eighth, and so on.† One of the chief characteristics of exponential decay is that, strictly speaking, the disintegration is not completely finished until after an infinite period of time. But since the quantity and hence also the radiation of UX sinks to about 10^{-3} of its original value after an interval of about 10×24 days, it follows that, for practical purposes, and provided it were initially not too strong, we can assume the preparation to have completely decayed after this time, i.e. the tenfold half-value period.‡ It is owing to this characteristic of exponential decay that we always consider the 'half-value period' (T) of a radio-element, or the time during which one-half of the amount initially available decays, instead of the absolute life period of the preparation. Although the latter quantity is always theoretically infinite, it is finite for practical purposes, but it depends on the strength of the preparation concerned and on the sensitiveness of our measuring instruments, whereas the half-value period does not.

Apart from the half-value period, we also make use of a quantity which is proportional to it, viz. the 'period of average life' (τ). This concept is defined here just as in insurance statistics. We multiply every life period by the average number of individuals that attain, but do not survive this period, and divide the sum of all these products by the total number of individuals. It can readily be shown mathematically that the period of average life (τ) so defined is equal to $\frac{1}{\lambda}$, the reciprocal of the disintegration constant λ, whereas the half-value period||

$$T = \frac{1}{\lambda}\log_e 2 = \frac{0{\cdot}69}{\lambda}.$$

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† Such a process is represented mathematically by the formula $N_t = N_0 e^{-\lambda t}$, where N_0 is the number of atoms originally present, and N_t the number present after a time t; λ is the disintegration constant, and e the base of the natural logarithms. It is assumed that the intensity of the measured radiation is always proportional to the number of atoms disintegrating per unit time.

‡ The radioactive substance can be detected for a longer period when we use more sensitive instruments towards the end of the measurement than at the beginning. The range of measurement over which one and the same instrument can be used rarely exceeds 1:1,000; hence by means of such an instrument the active preparation will be no longer detectable after a time of magnitude ten times the half-value period.

|| The half-value period (T) is the time for which $N_t = \frac{N_0}{2}$; and since $N_T = N_0 e^{-\lambda T}$, we

Thus, in the case of UX, the disintegration constant $\lambda = 0{\cdot}0283$ day^{-1}, and $\tau = 35{\cdot}4$ days, from which $T = 24{\cdot}5$ days.

The variation of the activity of UX separated from uranium, as a function of the time, is shown in curve *B* of Fig. 41, and this curve at the same time shows the proportional decrease in the number of UX atoms, on which the activity depends.

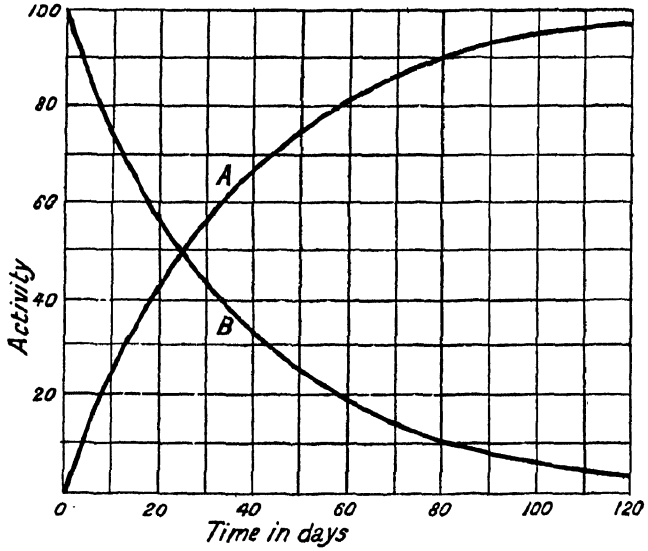

FIG. 41. Growth (*A*) and Decay (*B*) of Uranium X.

3. The Production of UX from Uranium

We shall now again turn our attention to the uranium preparation from which we have supposed the UX to have been removed, and which therefore no longer emits β-rays. It is found gradually to re-emit β-rays, and after the course of 24 (more exactly 24·5) days it exhibits just one-half of the β-radiation that it had before the separation of the UX. After 2 × 24 days it has reached $\frac{3}{4}$, after 3 × 24

must have $\frac{N_0}{2} = N_0 e^{-\lambda T}$. From this follows $T = \frac{1}{\lambda}\log_e 2$. The definition of the period of average life given above can be expressed mathematically in the form

$$\tau = \frac{\int\limits_0^\infty t\lambda N_0 e^{-\lambda t}\,dt}{N_0},$$

which is equal to $\frac{1}{\lambda}$. Since $\tau = \frac{1}{\lambda}$, we can also define the period of average life as the time within which the whole material would disintegrate, if the number of atoms disintegrating per unit time were to remain the same as initially: for

$$\frac{N_0}{N_0\lambda} = \frac{1}{\lambda} = \tau.$$

days $\frac{7}{8}$ of the value last referred to, and so on. Moreover, this rise also takes place exponentially, and is graphically represented by the curve *A* of Fig. 41. Just as the complete decay of the separated UX is to be expected theoretically only after an infinitely long period of time, but is for all practical purposes already attained after 10 × 24 days, so also the β-radiation of the UX-free uranium preparation will have practically reached its original strength after 10 × 24 days, although theoretically this should require an infinite period of time. In Fig. 41 the decay of the β-radiation of the separated UX and the growth of the β-radiation of the uranium preparation are represented on the same graph. From it we see that the sum of the two radiations, and hence the sum of the total number of UX atoms present, is constant at every instant, and equal to the initial value before the separation of the UX.

4. Radioactive Equilibrium

The disintegration theory enables us to predict the behaviour described above, as will be clear from the following example.

We shall suppose that we have 10^{15} atoms of uranium ($= 4 \times 10^{-7}$ gm.). Since the half-value period of uranium is approximately 10^{10} years, it follows that in 5 days, which we shall assume to be our unit of time, the relatively exceedingly small number of about 1,000 atoms will disintegrate, or only the 10^{12}th part of all the available atoms. In the next 5 days the 10^{12}th part of the now available atoms ($10^{15}-1{,}000$) will disintegrate; but the difference between this number and 10^{15} is so vanishingly small that, even if we require a very accurate calculation, we can assume them to be identical. Thus in each of the succeeding periods of 5 days' duration we shall repeatedly find that 1,000 atoms disintegrate. Not until after the lapse of many millions of years will the available amount of uranium have diminished appreciably, or the number of uranium atoms disintegrating in each period of 5 days be detectably different from 1,000, and this period of time is incomparably greater than the time involved in our investigation of the behaviour of the short-lived product UX.

Each 1,000 disintegrated atoms have ceased to be uranium; by virtue of the disintegration they are now UX atoms, but only for a short space of time. Since the period of average life of UX is only 35·4 days, about 15 per cent. or 150 of the 1,000 UX atoms will have already decayed after 5 days; of the remaining 850, 15 per cent. or 127 will decay in the next 5 days, and this number is markedly different from 150, in accordance with the corresponding difference between the numbers 850 and 1,000, and so on. Thus in the course of 5 days the process of disintegration exercises a very strong influence on the amount of UX present, and hence also on the amount disintegrating.

The activity is proportional to the amount disintegrating at any moment, and thus our previous discussion makes it clear why the activity of uranium is practically constant, whereas that of the separated UX diminishes by about 15 per cent. even after a period of 5 days.

Let us now consider how much uranium X would be formed if it were not separated from uranium, but left to itself. Whereas in each 5 days 1,000 fresh UX atoms are produced, independently of the number already present, the number of UX atoms disintegrating in the same time will be greater the greater the number that have already been formed. This behaviour necessarily leads to a state in which, for a given interval of time, the number of UX atoms disintegrating will be equal to the number freshly formed, so that the quantity of UX actually present no longer increases. This state is called 'radioactive equilibrium'. A radio-element is in a state of radioactive equilibrium with its disintegration product when the same number of atoms of the daughter element disintegrate as are formed in the unit of time. These important considerations will be quite clear from a study of Table 33.†

TABLE 33

Development of Radioactive Equilibrium

Number of time-intervals of 5 days	*Number of UX atoms produced in the interval by the decay of U atoms*	*Number of UX atoms available from the previous time-interval*	*Sum of the two numbers*	*Number of UX atoms decaying in the interval of time*	*Number of UX atoms remaining at the end of the time-interval*
1	1,000	..	1,000	150	850
2	1,000	850	1,850	277	1,573
3	1,000	1,573	2,573	386	2,187
4	1,000	2,187	3,187	478	2,709
5	1,000	2,709	3,709	556	3,153
6	1,000	3,153	4,153	623	3,530
7	1,000	3,530	4,530	680	3,850
8	1,000	3,850	4,850	728	4,122
9	1,000	4,122	5,122	768	4,354
10	1,000	4,354	5,354	803	4,551
..	....	....	....	...	....
..	....	....	....	...	....
..	....	....	....	...	....
50	1,000	5,667	6,667	1,000	5,667
51	1,000	5,667	6,667	1,000	5,667
52	1,000	5,667	6,667	1,000	5,667
53	1,000	5,667	6,667	1,000	5,667

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† The numbers in the table are not strictly correct, for during the time-unit of 5 days, which has been chosen on the grounds of clearness, the decay does not take place uniformly. A correct treatment presupposes that the time-unit can be neglected in comparison with the half-value period. The formulae of infinitesimal calculus operate with indefinitely small times, within which the decay is strictly constant.

We see from the table that the number of UX atoms disintegrating in the second interval of 5 days is almost double the number disintegrating in the first 5 days, whereas the difference between the numbers for the 10th and 9th intervals has already become small, and after about 50 time-intervals 1,000 atoms decay in 5 days, or, just as many as are produced. Thus the number of UX atoms remaining from the previous time-intervals remains constant at 5,667, or, in other words, radioactive equilibrium has been established. Fifty such time-intervals are 250 days, or approximately the tenfold value of the half-value period of UX. We have already seen that this multiple of the half-value period suffices for the attainment of the maximum β-activity of a uranium preparation that has been freed from its content of UX, i.e. for the establishment of radioactive equilibrium.

In determining how much UX, or another short-lived element, is present, it is usually sufficient to express the strength of the preparation in relative current units (e.g. 50 scale divisions per minute with a given electroscope). We may, however, wish to express how much by weight of the short-lived element is present, this being always formed in quantities that are too small to be weighed. This can be done most readily by making use of the conception of radioactive equilibrium. According to the basal assumption of the disintegration theory, the number of uranium atoms disintegrating per unit of time is equal to the product of the disintegration constant (λ_1) and the number of atoms available (N_1), or $N_1\lambda_1$. Similarly, for the number of UX atoms disintegrating per unit of time we obtain the product $N_2\lambda_2$, where N_2 is the number, and λ_2 the disintegration constant of the UX atoms. Now we have seen that in the state of radioactive equilibrium the number of UX atoms produced per unit of time is equal to the number disintegrating, and since the former number is equal to that of the uranium atoms disintegrating per unit of time, it follows that in the state of radioactive equilibrium

$$N_1\lambda_1 = N_2\lambda_2.$$

Since the weight of a single uranium atom is known (238 times the weight of an atom of hydrogen $= 238\times 1{\cdot}67\times 10^{-24} = 3{\cdot}97\times 10^{-22}$ gm.), we can evaluate N_1 by weighing the uranium. λ_1 and λ_2 (see p. 128) are known, and so we may directly obtain the required value of N_2. There are present, for example, in 1 gm. of uranium that has not been chemically treated for at least eight months

$$N_1 = \frac{1}{3{\cdot}97\times 10^{-22}} = 2{\cdot}52\times 10^{21} \text{ atoms},$$

$$\text{and}\quad N_2 = 2{\cdot}52\times 10^{21}\times\frac{\lambda_1}{\lambda_2} = \frac{2{\cdot}52\times 10^{21}\times 5{\cdot}0\times 10^{-18}}{3{\cdot}3\times 10^{-7}} = 3{\cdot}82\times 10^{10}.$$

Since 1 atom of UX weighs $234 \times 1{\cdot}67 \times 10^{-24} = 3{\cdot}91 \times 10^{-22}$ gm., the weight of the whole quantity of UX is equal to

$$3{\cdot}82 \times 10^{10} \times 3{\cdot}91 \times 10^{-22} = 1{\cdot}5 \times 10^{-11} \text{ gm.}^{\dagger}$$

It is possible to detect such a very small unweighable quantity with the aid of its β-radiation by virtue of the fact that UX disintegrates rapidly, and hence emits a relatively large number of β-rays. The determining factor for the detection of an element by radioactive means is not the quantity of it, but the product of the quantity and the disintegration constant.

5. Case in which the Life of the Parent Substance is not much longer than that of the Daughter Element. The Active Deposit of Thorium

Whereas the life period of uranium is so large that the quantity of it remains practically unaltered during the time required to establish radioactive equilibrium with its daughter element, in many cases, e.g. in the formation of ThC from ThB, the quantity of the parent substance varies appreciably during the production of the daughter element.

The transformation last mentioned also differs in other important points from that of uranium. Uranium is available in weighable quantities, and the separation of its transformation product is carried out by ordinary chemical methods; every method that is suitable for the separation of thorium from uranium is also applicable to the separation of UX (see p. 222). In the case of ThB and ThC, however, we have to do with a parent substance present in unweighable quantities, in the form of the 'active deposit', which has to be collected by a method quite new to chemistry.

As already mentioned (p. 1), one of the disintegration products of the radium, thorium, and actinium disintegration series is a gaseous substance called 'emanation'. The emanation of thorium (thoron) decays with a half-value period of 55 seconds, and the resulting atoms of thorium A and of its succeeding products are all metallic; the charged particles formed by the aggregation of such atoms are held in suspension in the air for some time, in consequence of their smallness, and are only deposited gradually on the surrounding surfaces. These disintegration products of the emanation further decay with appreciable rapidity, and are thus strongly active, and it is for this reason that the invisible deposit which they form on the surrounding surfaces is called the 'active deposit'.‡ As a consequence of their mode of production, most of

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† If we neglect the difference in atomic weight of the two elements, we can also obtain an approximately correct value for the equilibrium quantity by straightway inserting the amounts by weight, instead of the numbers of atoms N_1 and N_2.

‡ Since the active deposit exists on the surface of bodies in contact with which the

the particles carry a positive charge (cf. p. 70); this property supplies us with a means of preventing them from being deposited in the direction of the force of gravity. By introducing a negatively charged surface, e.g. a platinum wire at a potential of −100 volts, into the space in which the emanation decays, and in which the particles are in suspension, the active deposit can be collected upon it.

Fig. 42 represents an arrangement by means of which the active deposit may be collected.

The metallic base and the walls of the vessel are connected to the positive pole of a battery, and the platinum wire *W*, which passes through the insulating stopper of the vessel, is connected to the negative pole. If we introduce a thorium preparation *D* into the vessel, and do not remove the platinum wire until after the lapse of several days ('long exposure'), we find that it emits α-, β-, and γ-rays, and that its activity decreases with a half-value period of 10·6 hours, independently of which of the types of rays we may consider. This is illustrated by curve *B* in Fig. 43.†

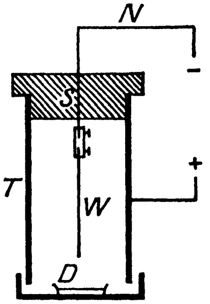

Fig. 42. Apparatus for collecting the Active Deposit.

Suppose we now repeat the experiment with only a 'short exposure' of a few minutes. We find that the wire now shows no initial α-activity after its removal from the activating vessel; but it gradually acquires an α-activity, the increase taking place in accordance with curve *A* in Fig. 43. After about 4 hours the curve reaches a maximum value, and the activity then decreases just like that for a long exposure, with a half-value period of 10·6 hours.

The most obvious explanation of this behaviour is that the substance ThB which is collected on the platinum wire during activation does not itself emit α-rays, but that these are emitted by the succeeding product ThC. The α-activity of the wire thus increases in the same degree as ThC is gradually formed from the disintegration of ThB. This process is continued until radioactive equilibrium has been established between the two substances, and thenceforward the variation with time of the activity of the platinum wire is just the same as if we had been dealing with the case of a long exposure.

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emanation has decayed in the form of an exceedingly fine and loose deposit, it can readily be transferred to other bodies, whereby they acquire radioactive 'contamination'. Before the material nature of the active deposit had been recognized, it was customary to call the activity acquired by bodies that had been in contact with emanation by the name 'induced activity'.

† The slower rate of decay of the activity indicated in curve *B* for the first hour or two is due to the fact that the state of 'transient equilibrium' between ThB and ThC is only gradually established (see p. 140).

The measurement of the β-activity of a platinum wire exposed to emanation for a short time lends support to the above conclusion, for the activity is present right from the beginning, which shows that ThB is a radio-element that emits β-rays.

From the fact that radioactive equilibrium between ThB and ThC is established after a few hours it can be concluded that the half-value period of the latter element is about one hour. The accurate value $T = 60{\cdot}5$ minutes can be calculated from the equation for the curve, or it may be obtained by the preparation of pure ThC and the determination of its decay curve. Thus, if we immerse a small nickel plate in a hydrochloric acid solution of the active deposit, it is found that ThC accumulates on the plate, and that it is practically free from ThB. By following the diminution in intensity of the α-activity of the separated ThC we obtain the same value for T as by the calculation referred to above.

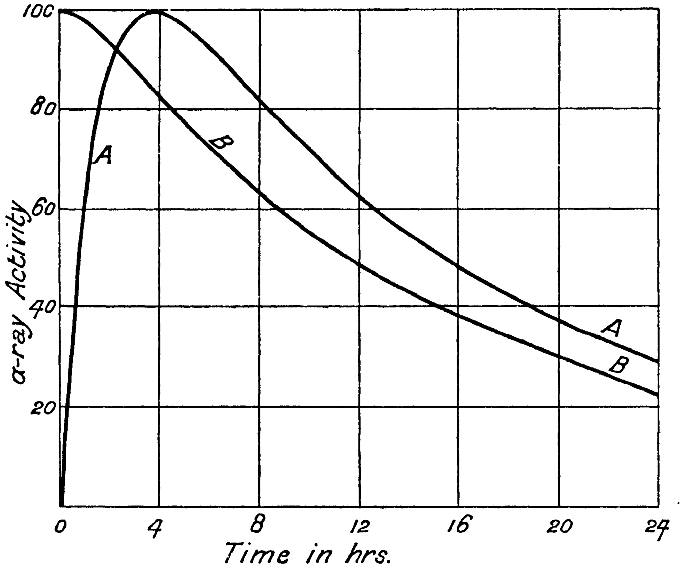

FIG. 43. Variation with Time of the Activity of the Active Deposit of Thorium, after a short (A) and after a long (B) exposure to thoron.

'Short' and 'long' exposures are limiting cases; for example, if we expose the platinum wire to emanation for an hour, the α-activity still increases initially, but instead of starting from zero it commences at a value intermediate between zero and the maximum value of the α-activity. In all cases, however, after radioactive equilibrium has been established for some time the activity diminishes with the half-value period of ThB. The parent substance of longer life gradually impresses its period on the daughter element.

Such a behaviour can be readily understood when we consider that in the state of equilibrium just as many ThC atoms disintegrate in unit time as are produced in the same time; but the latter number is also equal to the number of ThB atoms disintegrating, and since this is proportional to the total number of ThB atoms available, it follows that the number of ThC atoms disintegrating, and this determines the α-activity, will be proportional to the total number of atoms of its parent substance.

The behaviour just described is very often met with in radioactive analysis. When radon is in equilibrium with radium it disintegrates exceedingly slowly with the period of radium, although the half-value period of the radon is only 3·83 days. Polonium ($T = 140$ days) in equilibrium with RaD disintegrates with a half-value period of 22 years. When the half-value period of the daughter element is only slightly smaller than that of the parent element, the regular exponential decay with a half-value period equal to that of the parent element is not established until the lapse of a considerable period after the attainment of the maximum activity. For example, the activity of RaC ($T = 19{\cdot}7$ minutes) diminishes with a half-value period of more than 30 minutes for a protracted period after the maximum activity has been reached, instead of with the half-value period of RaB ($T = 26{\cdot}8$ minutes). However, since the behaviour of the active deposit of radium is on other grounds more complicated than that of the active deposit of thorium, we shall deal with it independently.

6. The Active Deposit of Short Life in the Case of Radium

Thorium emanation (thoron) is not directly transformed into ThB, for between these two elements there exists an intermediate product, ThA (see p. 144), which could be left out of the above discussion owing to its very short period ($T = 0{\cdot}14$ second). This cannot be done, however, in the case of the corresponding radium products. RaA has a half-value period of 3·05 minutes, a value which, compared with that of RaB (26·8 minutes) and RaC (19·7 minutes), is too great to be neglected. Thus the α-activity of a platinum wire that has been given a long exposure of about 5 hours in radon decays in the manner indicated by curve I in Fig. 44. From this we see that it decays rapidly at first, and then more and more nearly with the period of RaB. After a short exposure of only a few minutes, on the other hand, it decays very rapidly at first, with the period of RaA, as represented by curve II in Fig. 44. Then for some time it remains practically constant, for in this region the RaA has almost completely disintegrated; but almost in the same degree as the ionizing action of the RaA ceases the activity of the succeeding RaC increases, for it is being gradually formed and likewise emits α-rays. Before the RaC has attained equilibrium with the RaB, the RaA has completely disintegrated, and thenceforward similar conditions apply to those met with in the thorium series.

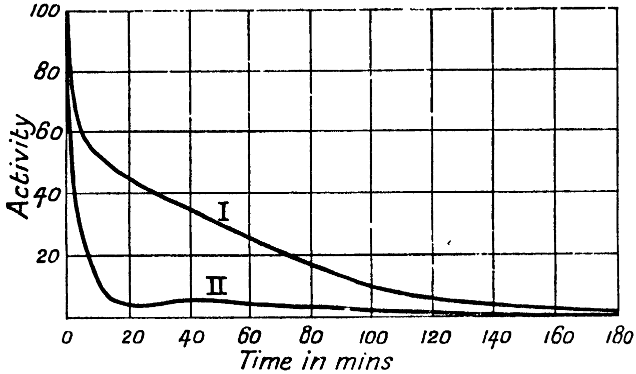

FIG. 44. Variation with Time of the Activity of the Active Deposit of Radium, after a long (I) and after a short (II) exposure to radon.

7. Production of an Element of Long Life from one of Short Life. The Long-Lived Active Deposit of Radium

Whereas the active deposit of thorium leads to a product which is at least practically stable or completely inactive, in the case of the active deposit of radium we can detect a residual activity even after the lapse of a few weeks, when the RaA, RaB, and RaC have completely disintegrated. Moreover, this activity continuously increases for a relatively long period of time. From the disintegration of the short-lived RaC the long-lived product RaD is formed, for which $T = 22$ years. The disintegration of this element leads by way of two further active products (RaE, $T = 5{\cdot}0$ days; and RaF, or polonium, $T = 140$ days) to the stable end-product RaG. The elements RaD, RaE, and RaF are also known collectively as the 'long-lived active deposit' of radium.[†]

RaD is a β-rayer the radiation from which can only be detected with difficulty, so that this is not suitable for the detection of the quantities of RaD available. On the other hand, the succeeding product RaE emits readily detectable β-rays, and they can be made use of for the identification and quantitative evaluation of the available quantities of RaD, in much the same way as we can utilize the α-radiation from ThC for the identification of its parent substance ThB. In the detection of the amount of RaD present we also frequently revert to the α-radiation

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[†] The reason why we can only gradually detect the long-lived active deposit of radium by electroscopic or photographic means is to be found in the extreme softness of the β-rays from RaD, which is almost inactive.

emitted by the third member of the long-lived active deposit, RaF, or polonium.[†]

If we collect the active deposit of thorium in the manner described, we shall arrive at the practical maximum of activity attainable in about 3 days, and in the corresponding case of the short-lived active deposit of radium in as short a period as 5 hours. But we should have to collect the long-lived active deposit of radium for over 100 years in order to reach its greatest possible activity. During this time, however, the quantity of radium in our activating vessel, which is the source of the radon and of the active deposit, will have diminished, whereas we have been able to regard it hitherto as constant ($T = 1{,}590$ years). In consequence of this it can be shown that after the lapse of 110 years the number of RaD atoms resulting from a given quantity of radium will have passed its maximum value.

It follows from the above considerations that, when we prepare the strongest RaC preparation possible after an exposure of several hours, we shall by no means obtain a strong RaD (or RaF) preparation as a result of its decay. Large quantities of RaD can be accumulated only by the utilization of extended intervals of time, as on the walls of a closed vessel in which a large quantity of radon has disintegrated, or in radium preparations sealed off in glass tubes, or in minerals and the like. The quantity of RaD in equilibrium with 1 gm. of radium amounts to about 13 mg.

[†] The element RaE, which comes between RaD and RaF, can often be left out of account in these measurements, owing to its relatively short half-value period.

REFERENCES

Detailed accounts of the topics embodied in this chapter will be found in *Radioactive Substances and their Radiations*, by E. RUTHERFORD (Cambridge University Press, 1913), in *Radioaktivität*, by ST. MEYER and E. VON SCHWEIDLER (Teubner, Leipzig, 1927), and in *Radioactivité*, by Mme P. CURIE (Hermann and Co., Paris, 1935).

XII

MATHEMATICAL TREATMENT OF DISINTEGRATION PROCESSES

ALTHOUGH the analysis of radioactive processes can sometimes be performed graphically, in most cases we have to revert to calculation. We shall therefore briefly discuss in this chapter the mathematical treatment of a few of the more important disintegration processes.

1. Disintegration of a Pure Substance

According to the fundamental assumption of the disintegration theory, the number of atoms disintegrating in unit time is proportional to the number of atoms available at the instant considered. From this it immediately follows that, if N_0 atoms are present initially, only $N_t = N_0 e^{-\lambda t}$ atoms will be available after a time t, where λ is the disintegration constant and e the base of natural logarithms (cf. p. 127).

Example. Suppose that there are 1,000 relative units of ThB originally present (e.g. scale divisions per minute, measured with an α-ray electroscope). The number of units remaining after the lapse of one hour ($=$ 3,600 seconds) will be $1{,}000 \times e^{-1\cdot82\times10^{-5}\times3{,}600} = 937$, since $\lambda = 1\cdot82 \times 10^{-5}$ sec.$^{-1}$

2. Production of a Pure Substance from a Parent Element Constant in Amount

If we wish to calculate the growth of the daughter element, or the fraction of the maximum quantity N_∞ obtainable, after the lapse of the time t, we apply the formula:

$$N_t = N_\infty(1-e^{-\lambda t}).$$

This expression follows immediately from the reciprocal nature of the growth and decay curves (cf. p. 128).

Example. Suppose we have mesothorium in equilibrium with all its disintegration products up to and including ThA, so that we may regard the immediate parent of ThB as being constant in amount; then if the maximum quantity of ThB obtainable from this preparation after an 'infinitely' long exposure amounts to 1,000 relative units, we should have, after an exposure of one hour,

$$N_t = 1{,}000(1-e^{-1\cdot82\times10^{-5}\times3{,}600}) = 63 \text{ units.}$$

3. From a Substance I, of which there are initially ($t = 0$) A_0 Atoms present, the Elements II, III, etc., are successively formed

We shall represent the numbers of atoms of the elements II, III, etc., by the letters B, C, etc. We require to find the magnitudes of A, B, C, etc., at a particular instant of time. Suppose we expose a platinum wire in radium emanation (radon) for a short time, during which practically only RaA is deposited. We measure its initial activity (= amount of RaA), and calculate how many of the RaB and RaC atoms formed from the RaA will be present after the lapse of the time t. The number of atoms of RaB will be

$$B = A_0 \frac{\lambda_A}{\lambda_B - \lambda_A}\{e^{-\lambda_B t} - e^{-\lambda_A t}\},$$

and of RaC,

$$C = A_0\left[\left\{\frac{\lambda_A \lambda_B}{(\lambda_B-\lambda_A)(\lambda_C-\lambda_A)}\right\}e^{-\lambda_A t} + \left\{\frac{\lambda_A \lambda_B}{(\lambda_C-\lambda_B)(\lambda_A-\lambda_B)}\right\}e^{-\lambda_B t} + \left\{\frac{\lambda_A \lambda_B}{(\lambda_A-\lambda_C)(\lambda_B-\lambda_C)}\right\}e^{-\lambda_C t}\right],$$

where λ_A, λ_B, and λ_C are the relevant disintegration constants, and A_0 denotes the original number of atoms of RaA present.

Thus, if at the time $t = 0$ there were 1,000 atoms of RaA present, then after 10 minutes ($t = 600$ sec.) there would be 103 atoms of RaA, 756 atoms of RaB, and 124 atoms of RaC; the remaining 17 atoms would already have been transformed into RaD.

4. At the Time $t = 0$ we have Radioactive Equilibrium between the Transformation Products I, II, III, etc.

How many atoms A, B, C, etc., will be present after a definite time t? This case corresponds to a long exposure in radon, and we require to calculate the number of atoms of RaA, RaB, and RaC still remaining on the platinum wire t seconds after its removal from the radon.

Since radioactive equilibrium exists between the several products at the time $t = 0$, we must have

$$\lambda_A A_0 = \lambda_B B_0 = \lambda_C C_0 = n_0.$$

Thus if there were 1,000 atoms of RaA present initially ($A_0 = 1{,}000$), the initial numbers of atoms of RaB and RaC would be 8,829 and 6,395 respectively.

At the time t the following numbers of atoms will be available:

$$A = \frac{n_0}{\lambda_A} e^{-\lambda_A t},$$

$$B = \frac{n_0}{\lambda_A - \lambda_B}\left[\frac{\lambda_A}{\lambda_B}e^{-\lambda_B t} - e^{-\lambda_A t}\right],$$

$$C = n_0(a.e^{-\lambda_A t} + b.e^{-\lambda_B t} + c.e^{-\lambda_C t}),$$

where

$$a = \frac{\lambda_B}{(\lambda_B - \lambda_A)(\lambda_C - \lambda_A)}, \quad b = \frac{\lambda_A}{(\lambda_A - \lambda_B)(\lambda_C - \lambda_B)}, \quad c = \frac{\lambda_A \lambda_B}{\lambda_C(\lambda_A - \lambda_C)(\lambda_B - \lambda_C)};$$

thus after 10 minutes we should have

$$A = 103, \quad B = 7{,}543, \quad C = 6{,}276.$$

5. 'Secular' and 'Transient' Radioactive Equilibrium

In the previous example the equilibrium quantities of RaA, RaB, and RaC have been calculated on the assumption that the radon, from which the RaA is formed, remains constant in amount, i.e. is itself in equilibrium with radium. When this is not the case, as when the exposure is made in a vessel that contains only radon, we are no longer justified, strictly speaking, in neglecting the rate of decay of the radon as compared with that of the short-lived active deposit. It is true that even here we obtain a state of equilibrium between the four products, but the relative quantities involved are somewhat different. The balance is in favour of the later products, the decay of which cannot keep pace with that of the parent substance, so that it is not possible to attain 'secular' equilibrium. This state, which is very often met with in practice, is called 'transient' equilibrium. For instance, in the case of radon, the quantities of RaA, RaB, and RaC corresponding to transient equilibrium are respectively about 0·05 per cent., 0·5 per cent., and 1 per cent. greater than those corresponding to the state of secular equilibrium. These numbers are calculated from the formulae:

for RaA $\dfrac{\lambda_A}{\lambda_A - \lambda_{Rn}}$,

for RaB $\dfrac{\lambda_A \lambda_B}{(\lambda_A - \lambda_{Rn})(\lambda_B - \lambda_{Rn})}$,

for RaC $\dfrac{\lambda_A \lambda_B \lambda_C}{(\lambda_A - \lambda_{Rn})(\lambda_B - \lambda_{Rn})(\lambda_C - \lambda_{Rn})}$.

From these formulae we see that the ratio of the equilibrium quantities corresponding respectively to transient and secular equilibrium will become unity in each case when the rate of decay of the parent substance can be neglected. The nearer this rate of decay approaches to those of the succeeding products, on the other hand, the greater is the value of the ratio. Thus when we are dealing with the decay of ThX which is not in equilibrium with its long-lived parent element radiothorium, the correction for the succeeding products ThB, ThC, and ThD already amounts to from 13 to 15 per cent.

REFERENCES

In connexion with this chapter reference should be made to *Radioactive Substances and their Radiations*, by E. RUTHERFORD (Cambridge University Press, 1913), to *Radioaktivität*, by ST. MEYER and E. VON SCHWEIDLER (Teubner, Leipzig, 1927), and to *Radioactivité*, by Mme P. CURIE (Hermann & Co., Paris, 1935).

XIII

THE THREE TRANSFORMATION SERIES

THE investigation of the radioactive transformations on the basis of the disintegration theory leads to the result that all natural radioactive substances, with the exception of potassium, rubidium, and samarium, the activity of which elements still demands further investigation, are products of disintegration of one or other of the parent substances uranium and thorium. A schematic representation of the successive products of the uranium, actinium, and thorium families respectively is given in Tables 34 and 35.

In these tables a radio-element is always placed lower down in the table than its parent substance when its production has involved the emission of α-radiation; in this case its atomic weight is always four units less than that of the parent substance, and this fact is expressed in the left-hand column. If the daughter element is the result of the emission of β-radiation by the parent element, so that its atomic weight is practically unchanged, it is placed to the right alongside its parent element.

In certain cases two arrows branch off from the parent element, indicating that this substance is transformed into two different new products. Thus we see that of the two products that are formed respectively from RaC, AcC, and ThC, one is produced by the emission of α-radiation, and the other by the emission of β-radiation. Such branching in the disintegration process is known as 'dual decay'. The 'branching ratio' may differ greatly in different cases. Thus from 10,000 RaC atoms only four are transformed into RaC″, whereas 9,996 go to form RaC′; the reverse holds for AcC, for which the majority of the atoms are transformed into AcC″ (about 9,984 in 10,000); finally, with ThC the difference is less marked, for 65 per cent. is transformed into ThC′ and 35 per cent. into ThC″, and both branches probably lead to the same stable isotope of lead, thorium D.

The observation of such relatively insignificant branches as those of RaC″ and AcC′ suggests the possibility that there may be other places in the disintegration series at which multiple disintegration may take place. Thus the radio-element UZ (see p. 222) has usually been regarded as a branch product of the uranium series; but recent work seems to render it more probable that UZ is actually a product of disintegration of an isotope of uranium.

It might be expected that the energy liberated in the transition from the C- to the D-products of disintegration would differ according to which of the two branches is traversed, i.e. $\mathrm{C} \xrightarrow{\beta} \mathrm{C}' \xrightarrow{\alpha} \mathrm{D}$, or

Table 34

Uranium-Radium and Uranium-Actinium Disintegration Series

$$\begin{tikzcd}
\textit{Atomic Weight} & & & & & & & \\
238 & \text{Uranium I} \arrow[d] & & & & & & \\
234 & \text{Uranium X}_1 \arrow[r] & \text{Uranium X}_2 \arrow[r] & \text{Uranium II} \arrow[d] & & & & \\
230 & & & \text{Ionium} \arrow[d] & & & & \\
226 & & & \text{Radium} \arrow[d] & & & & \\
222 & & & \text{Radon} \arrow[d] & & & & \\
218 & & & \text{RaA} \arrow[d] & & & & \\
214 & & & \text{RaB} \arrow[r] & \text{RaC} \arrow[r] \arrow[d] & \text{RaC}' \arrow[d] & & \\
210 & & & & \text{RaC}'' \arrow[r] & \text{RaD} \arrow[r] & \text{RaE} \arrow[r] & \text{RaF} \arrow[d] \\
206 & & & & & & & \text{RaG (Stable lead isotope)}
\end{tikzcd}$$

$$\begin{tikzcd}
235 & \text{Actino-Uranium} \arrow[d] & & & & \\
231 & \text{Uranium Y} \arrow[r] & \text{Proto-actinium} \arrow[d] & & & \\
227 & & \text{Actinium} \arrow[r] & \text{Radio-actinium} \arrow[d] & & \\
223 & & & \text{Actinium X} \arrow[d] & & \\
219 & & & \text{Actinon} \arrow[d] & & \\
215 & & & \text{AcA} \arrow[d] & & \\
211 & & & \text{AcB} \arrow[r] & \text{AcC} \arrow[r] \arrow[d] & \text{AcC}' \arrow[d] \\
207 & & & & \text{AcC}'' \arrow[r] & \text{AcD (Stable lead isotope)}
\end{tikzcd}$$

$C \xrightarrow{\alpha} C'' \xrightarrow{\beta} D$. But if the maximum observed β-energy, corresponding to the upper limit of the continuous β-spectrum, be inserted in the energy equation, it is found that the energy changes along the two paths are identical. Thus for the radium disintegration series we have:

$$\begin{array}{llll}
\text{RaC} \xrightarrow{\beta} \text{RaC}' = 3{\cdot}1 \times 10^6 \text{ e.V.} & & \text{RaC} \xrightarrow{\alpha} \text{RaC}'' = 5{\cdot}4 \times 10^6 \text{ e.V.} \\
\text{RaC}' \xrightarrow{\alpha} \text{RaD} = \underline{7{\cdot}7 \times 10^6 \text{ e.V.}} & & \text{RaC}'' \xrightarrow{\beta} \text{RaD} = \underline{5{\cdot}0 \times 10^6 \text{ e.V.}} \\
\qquad\qquad\quad 10{\cdot}8 \times 10^6 \text{ e.V.} & & \qquad\qquad\quad 10{\cdot}4 \times 10^6 \text{ e.V.}
\end{array}$$

TABLE 35

Thorium Disintegration Series

Atomic Weight	
232	Thorium
	↓
228	Meso-thorium 1→Meso-thorium 2→Radiothorium
	↓
224	Thorium X
	↓
220	Thoron
	↓
216	ThA
	↓
212	ThB→ThC→ThC′
	↓ ↓
208	ThC″→ThD (Stable lead isotope)

Similar agreement is found also for the branch products of the actinium disintegration series, and to a lesser degree for those of the thorium series, but not for the β-branching of UX_1, which suggests that UZ is not a branch product of the disintegration (see above). From these results it seems probable that the D-product formed in each branching process is independent of the branch traversed.

It should be mentioned in connexion with the uranium-actinium series that the identity of the parent substance and the atomic weights of the products are somewhat uncertain. In ordinary lead an isotope of mass 207 is always found, and since this originates neither from the radium nor from the thorium disintegration series it is assumed that this type of lead is identical with the end-product AcD. The atomic weights of the various products of the actinium series in the above table have been calculated on this basis. The recent discovery of an isotope of uranium of mass 235, presumably actino-uranium, gives added support to the data embodied in the table.

REFERENCES

Detailed discussion of the subject-matter of this chapter will be found in *Radioactive Substances and their Radiations*, by E. RUTHERFORD (Cambridge University Press, 1913), and in *Radioaktivität*, by ST. MEYER and E. VON SCHWEIDLER (Teubner, Leipzig, 1927).

XIV

DETERMINATION OF VERY LONG AND OF VERY SHORT HALF-VALUE PERIODS

THE half-value periods of the radio-elements vary between very wide limits ranging from intervals of time that are inconceivably small (e.g. for ThC′, $T = 10^{-11}$ sec.) to periods that are literally enormous (e.g. for thorium, $T = 1{\cdot}34\times 10^{10}$ years). Although the determination of these values cannot be carried out directly, they can nevertheless be evaluated in a relatively simple manner, and in many cases it is possible to determine them in several independent ways.

Suppose we consider the case of uranium I. If this element is in equilibrium with its later disintegration product radium, as is always the case in minerals of great geological age, then (p. 131) the product of the number of available atoms and the disintegration constant for uranium I is equal to the corresponding product for radium,

$$\mathrm{U_I}\times\lambda_{\mathrm{U_I}} = \mathrm{Ra}\times\lambda_{\mathrm{Ra}},$$

or if the amounts of the elements be expressed in grams, whereby we must take account of the ratio of the atomic weights (238/226),

$$\mathrm{U_I}\times\lambda_{\mathrm{U_I}} = \frac{238}{226}\times\mathrm{Ra}\times\lambda_{\mathrm{Ra}}.$$

The ratio of the radium and uranium contents of old minerals has been established by numerous chemical analyses to be $3{\cdot}4\times 10^{-7}$,[†] and since $\lambda_{\mathrm{Ra}} = 1{\cdot}38\times 10^{-11}$ sec.$^{-1}$, it follows that

$$\lambda_{\mathrm{U_I}} = 4{\cdot}9\times 10^{-18}\ \text{sec.}^{-1},$$

and

$$T = 4{\cdot}4\times 10^{9}\ \text{years}.$$

Another method which leads to the evaluation of the required disintegration constant is the counting of the number of α-particles emitted per second by a known quantity of uranium. Since it has been established that each disintegrating atom ejects only one α-particle (p. 21), it follows that the number of ejected α-particles, and these can be counted, must be equal to the number of atoms disintegrating. Moreover, the disintegration constant is equal to the ratio of the number of atoms disintegrating per unit of time to the total number of atoms of a radio-element present (p. 126). Now 1 gram ($= 2{\cdot}52\times 10^{21}$ atoms)

[†] The value found for uranium also holds for uranium I, since the amount by weight of uranium II present in uranium is only 0·008 per cent. (see p. 221).

of uranium emits $2{\cdot}4 \times 10^4$ α-particles per second, half of these being due to uranium I and the other half to uranium II. From this we have

$$\lambda_{\mathrm{U_I}} = \frac{1{\cdot}2 \times 10^4}{2{\cdot}52 \times 10^{21}} = 4{\cdot}76 \times 10^{-18} \text{ sec.}^{-1},$$

and $$T_{\mathrm{U_I}} = 4{\cdot}6 \times 10^9 \text{ years.}$$

This value is in good agreement with that found by the method previously discussed.

Relation between the Disintegration Constant and the Velocity of the Rays Emitted

A simple empirical relation has been found to exist between the disintegration constant and the range (or velocity, see p. 21 et seq.) for radio-elements that emit α-rays. By virtue of this relation we are enabled by extrapolation to derive a rough estimate of the disintegration constant even for elements of extremely short life. If, as in Fig. 37 (see p. 106), we plot the logarithm of the range as abscissa and the logarithm of the disintegration constant as ordinate, we obtain a straight line for each of the three disintegration series. The range corresponding to a definite disintegration constant is greater for the actinium series than for the thorium series, whilst that for the latter series is greater than the range for the uranium series.†

Thus by utilizing Fig. 37 we obtain the value 10^{11} sec.$^{-1}$ for the disintegration constant of ThC′ from the known range 8·6 cm. at 15° C. This corresponds to a half-value period of 10^{-11} seconds.

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† The following relation between the disintegration constant and the velocity of the α-particles emitted can be applied quite generally to all α-rayers:

$$\log\lambda = 20{\cdot}46 - 1{\cdot}191 . 10^9 . \frac{Z-2}{v} + 4{\cdot}08 . 10^6 . \sqrt{(Z-2)} . \sqrt{r},$$

where λ = disintegration constant (sec.$^{-1}$); r = nuclear radius; Z = atomic number; v = velocity of the α-particles. Since the α-particle velocity is proportional to $\sqrt[3]{\text{range}}$, it follows that the above relation also connects the disintegration constant and the range of the α-particles emitted (cf. p. 105).

XV

CHEMICAL NATURE AND ISOTOPY OF THE RADIO-ELEMENTS

ONLY a few of the radioactive elements, e.g. thorium, uranium, and radium, can be obtained in weighable quantities. The determination of the properties of these elements is carried out by means of the usual chemical methods, for the radioactive nature of the element does not affect its chemical properties, and an atom of radium, to take one example, is during the period of its stability quite analogous to the atoms of the typical metals of the alkaline earths. From the moment of its disintegration, however, we are no longer dealing with an atom of radium, but with an atom of a new element—the emanation (radon) produced from radium—which, so long as it lasts, behaves completely like the atoms of the other inert gases helium, neon, etc. (See also footnote, p. 161.)

The large majority of the radio-elements can only be obtained in unweighable quantities, and in many cases the amounts of them available are quite invisible. How then was it possible to obtain information on the chemical nature of these substances?

The principle of the method adopted consisted in adding a known element (*A*) to the solution containing infinitesimal traces of the radio-element to be investigated, and then precipitating it with a suitable reagent after the solution had been thoroughly mixed. After filtration an electroscopic examination of the precipitate and of the solution after evaporation to dryness revealed whether or not the radio-element had been carried down by the element *A* in the above operation. If it had, the precipitate was again dissolved, and another element (*B*) of similar chemical properties was added, after which a separation of *A* and *B* was undertaken. In this way it would be found with which of the two substances *A* or *B* the radio-element was separated, and hence, from the viewpoint of analytical chemistry, to which of them it had the greater resemblance. By a continuation of this process it was always possible to find the element most nearly resembling the unknown radio-element in its chemical reactions. It is necessary to emphasize, however, that the conclusions which were drawn about the chemical nature of a radio-element from this 'co-precipitation' process did not carry conviction until it was proved that non-specific adsorption forces do not play a part in this process. In Chapter XVII we shall deal with the rules which apply to the behaviour of such small quantities of a substance; in point of fact, they were first disclosed by the study of the radio-elements.

Now the above described process of finding for every radio-element that known element which most closely resembles it has in many cases elicited a surprisingly simple result. With very many of the radio-elements it has been found, for instance, that their resemblance to a particular element is so great that, after admixture of this element, it has not been possible by any process to separate them again. It has in fact proved impossible, by fractional crystallization of this element, to achieve even the slightest increase in concentration of the radio-element in a particular fraction. The radio-element and the admixed inactive element behaved completely like a single chemical individual.

The phenomenon could only be explained by what was then quite a novel assumption, that the atoms of the radio-element have the same chemical properties as those of the inactive element, but that the two differ from each other in atomic weight and radioactive behaviour. On this theory the radio-element was only a particular type of the known element; from the chemical point of view it was not a new element, but belonged to the same place in the periodic classification of the elements. For this reason it was called an 'isotope' of the more common element.

This phenomenon of isotopy was discovered simultaneously also in the field of the inactive elements. To-day we know that the majority of the ordinary elements consist of more than of one kind of atom, and we shall have more to say on the phenomenon of isotopy for common elements in Chapter XIX. For them the difference between the isotopic types of atom of one and the same element lies only in their weights. For this reason alone the phenomenon of isotopy among the radio-elements is of a unique and particularly interesting kind, for here the various atomic types differ not only in their weights but also in their different ability to disintegrate. Moreover, in the field of the ordinary elements we always find a uniform mixture of the isotopic types of atoms, whereas with the radio-elements, although they sometimes occur in association with their inactive isotopes, as in the cases of radium D and ionium, in the vast majority of cases they can also be prepared free from their inactive isotopes. Because of this fact, in particular, the radio-elements have been found suitable for the study of the essential properties of isotopes, and in what follows we shall discuss a few of these investigations in greater detail.

If, for instance, we endeavour to separate or to concentrate the RaD contained in lead chloride derived from pitchblende, we find that we are no nearer our goal after numerous crystallizations of known lead compounds, or by partial sublimation of these compounds or of the metallic lead. Other methods are equally fruitless, such as electrolysis in aqueous solution or in the fused state, adsorption by charcoal or other substances, diffusion, dialysis, and so on. The difficulty of separating closely related substances like those of the rare earths is a phenomenon well known in chemistry, but the difficulty of separation in such cases is not to be compared with that confronted when we are dealing with isotopes. We can always appreciably displace the concentration ratio in a mixture of the rare earths by means of various chemical operations, but this is not possible with isotopes unless we have recourse to special methods (cf. Chap. XX), although the quantitative determination of the ratio of the constituents, especially for the rare earths, is often faced with great difficulties, quite unknown in radiochemistry. Thus we can find the RaD-content of a lead-RaD mixture in a most convenient and accurate manner by means of an electroscope, and using the α-radiation of its succeeding product polonium (see p. 136).

If we do not proceed from a natural mixture of RaD and lead such as is found in minerals, but prepare a mixture artificially by mixing a pure RaD salt, obtained from the decay of the gaseous emanation, with a lead salt, we find that this mixture likewise shows the behaviour described above.

In consequence of the ease with which it can be detected, RaD is particularly suitable for the control of the concentration ratio of a mixture of lead isotopes before and after the application of chemical operations. Nevertheless, in the investigation of the properties of an unmixed isotope—e.g. its spectral behaviour, the absolute solubility of its salts, its density, its normal electrochemical potential, etc., and above all its atomic weight—the stable end-product of the uranium series RaG is to be preferred, for although it is inactive, it is an isotope of lead that can be obtained in much greater quantities.

Pure RaG is obtained from uranium minerals that have not been contaminated by common lead in the course of geological time, for example, from a crystalline uraninite from East Africa. Owing to the smallness of the quantity of RaD that can be obtained from radium emanation, it is not possible to carry out an atomic weight determination of this isotope of lead, and our conclusions as to the difference of the atomic weights of RaD and common lead (atomic weight = 207·21) are based solely on the formula

$$\underset{226}{(\text{At. wt. of Ra})} - 4\times\underset{16}{(\text{At. wt. of He})} = \underset{210}{(\text{At. wt. of RaD})}.$$

But in the case of RaG it has been possible to establish experimentally that its atomic weight is different from that for common lead. The value 206·03 was found for the atomic weight of RaG, whereas the value predicted by calculation† was

$$\underset{226}{(\text{At. wt. of Ra})} - 5\times\underset{20}{(\text{At. wt. of He})} = \underset{206}{(\text{At. wt. of RaG})}.$$

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† In exact calculations of this kind the decrease in mass due to the loss of energy has to be taken into account. In the gradual transformation of uranium into lead,

Most uranium minerals, e.g. pitchblende from Joachimstal, contain ordinary lead in addition to numerous other mineralogical impurities, and under the most favourable conditions we obtain from this mineral a mixture of about 36 per cent. of lead with 64 per cent. of RaG, with a 'combining weight' of 206·4. On the other hand, the mineral curite (containing 21·3 per cent. PbO and 74·2 per cent. UO_3), which occurs in large quantity at Katanga (Belgian Congo), contains no trace of ordinary lead, and the lead extracted from it is pure radium G.

The specific gravity of metallic RaG is related to that of common lead in the same ratio as their atomic weights, from which it follows that the atomic volumes of the two elements are equal. This is shown in Table 36.

TABLE 36

Atomic Volumes of Different Types of Lead

Type of lead	*Atomic weight*	*Specific gravity*	*Atomic volume*
Ordinary lead. .	207·21	11·337	18·28
Uranium-lead (RaG)	206·03	11·273	18·28

From the fact that the density of a saturated solution of RaG nitrate in water is correspondingly smaller than that of ordinary lead nitrate in water it follows that the molar solubilities of these two substances are equal (see Table 37). Moreover, no difference has been detected in the refractive indices of the saturated solutions, as is also indicated in Table 37.

TABLE 37

Saturated Aqueous Solutions of the Different Types of Lead

Salt	*Molar concentration*	*Refractive index*
Ordinary lead nitrate .	1·7993	1·7815
Uranium-lead nitrate .	1·7991	1·7814

The normal electrochemical potential of RaG in a solution of $RaG(NO_3)_2$ is at least within 1/10 milli-volt identical with that of lead in a solution of $Pb(NO_3)_2$. The mobility of these two ions is identical within

8 α-rays and 6 β-rays are emitted, with a total kinetic energy of 52×10^6 e.V., which represents a loss of mass of 0·056 (see Table 57). We should expect, therefore, that the atomic weight of pure RaG would be

$$238{\cdot}07_4 - 8 \times 4{\cdot}002 - 0{\cdot}056 = 206{\cdot}00_2,$$

If allowance be made for the probable percentage content of actino-uranium (^{235}U) in uranium (c. 1%), the atomic weight of U_1 becomes $238{\cdot}10_5$, and that of RaG becomes $206{\cdot}03_3$ [1].

1/100 per cent. The X-ray spectra are also the same to a degree of accuracy of about 1/200 per cent. of the wave-length, whereas the optical spectrum reveals only exceedingly small differences. The melting-points do not differ by more than 0·06° C. The temperature of commencement of superconductivity for ordinary lead (7·2° A.) was found to be identical within 1/40° with that at which superconductivity begins with uranium-lead. Moreover, no difference could be detected in the form of the resistance curves between 7·2° and 15° A. Thus for two isotopic mixtures of mercury obtained artificially and differing in combining weight by 0·1 of a unit, the resistance was found to be the same to within $1:10^6$.

Still another isotope of lead, ThD, the end-product of the thorium series, is obtainable in weighable quantities. It should possess an atomic weight of $232-(6\times4) = 208$. Unfortunately, the large majority of thorium minerals also contain uranium (sometimes also common lead), and hence we obtain from these minerals a mixture of RaG with ThD, in consequence of which the atomic weight appears to be greatly reduced. The purest ThD hitherto obtained was separated from a thorite from Norway containing practically no uranium, and it yielded a combining weight of 207·9.

Of the remaining isotopes, ionium is the only one (excepting protoactinium) that can be obtained in weighable quantities. It is isotopic with thorium. Io-Th mixtures prepared from Joachimstal pitchblende and having an ionium content of *c.* 30 per cent. possess a combining weight of 231·51, whereas thorium has an atomic weight of 232·12.

All the known isotopes of radioactive elements are listed together in Table 38, which also contains the atomic weights and half-value periods of the substances concerned. The grouping of the elements was carried out in most cases on the basis of chemical experiments described at the beginning of this chapter, namely, by establishing their inseparability from known substances. In the case of the substances of shortest life recourse had to be made to the radioactive 'displacement law' discussed in the next chapter. Each group is arranged in the order of diminishing half-value period, and begins with the member of longest life, from which, in general, it receives its name. Thus we speak of 'types of lead', 'types of thorium', and so on. We notice that the table, in which the elements gold and mercury are included for purposes of completeness, embraces only the last two horizontal rows of the periodic classification (cf. Tables 40 and 41, pp. 158, 159, which present the entire periodic system of the elements). The isotopes of mercury and of other non-radioactive substances are referred to in Chapter XIX.

TABLE

Radioactive Isotopes (with Atomic

Period	*Group I*	*Group II*	*Group III*	*Group IV*
VI	79	80	81	82
	Au 197·2	Hg 200·61	Tl 204·39	Pb 207·21
	stable	stable	stable	stable
			AcC″ 207	RaG 206
			4·76 minutes	stable
			ThC″ 208	ThD 208
			3·1 minutes	stable
			RaC″ 210	AcD 207
			1·32 minutes	stable (?)
				RaD 210
				22 years
				ThB 212
				10·6 hours
				AcB 211
				36·0 minutes
				RaB 214
				26·8 minutes
VII	87	88	89	90
		Ra 226·0	Ac 227	Th 232·12
		1,590 years	13·5 years	$1{\cdot}34 \times 10^{10}$ years
		$MsTh_1$ 228	$MsTh_2$ 228	Io 230
		6·7 years	6·13 hours	$8{\cdot}3 \times 10^{4}$ years
		AcX 223		RdTh 228
		11·2 days		1·90 years
		ThX 224		UX_1 234
		3·64 days		24·5 days
				RdAc 227
				18·9 days
				UY 231
				24·6 hours

38

Weights and Half-Value Periods)

Group V	*Group VI*	*Group VII*	*Group VIII* (0)
83 Bi 209·00 stable RaE 210 5·0 days ThC 212 60·5 minutes RaC 214 19·7 minutes AcC 211 2·16 minutes	84 Po 210 140 days RaA 218 3·05 minutes ThA 216 0·14 seconds AcA 215 0·002 seconds AcC′ 211 5×10^{-3} seconds RaC′ 214 10^{-6} seconds ThC′ 212 10^{-11} seconds	85	86 Rn 222 3·825 days Tn 220 54·5 seconds An 219 3·9 seconds
91 Pa 231 $3{\cdot}2\times10^{4}$ years UZ 234 6·7 hours UX$_2$ 234 1·14 minutes	92 U$_{\text{I}}$ 238·07 $4{\cdot}4\times10^{9}$ years AcU 235 *c.* 7×10^{8} years U$_{\text{II}}$ 234 $3{\cdot}4\times10^{5}$ years		

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XVI

THE PLACE OF THE RADIO-ELEMENTS IN THE PERIODIC CLASSIFICATION OF THE ELEMENTS. THE DISPLACEMENT LAW

THE discovery of the large number of about forty radioactive substances brought chemists face to face with the problem as to the possibility of accommodating these substances in the periodic classification of the elements. A method for the rational grouping of the radio-elements suggests itself, as revealed in Table 38, when we take the phenomenon of isotopy as the basis of our considerations. According to this, most of the radio-elements are chemically inseparable from other radioactive elements or from stable elements. If we adhere to the principle of assigning isotopes to one and the same place in the periodic classification, it is found that from uranium to thallium the number of available places in the classification suffices for the accommodation of all the radio-elements.

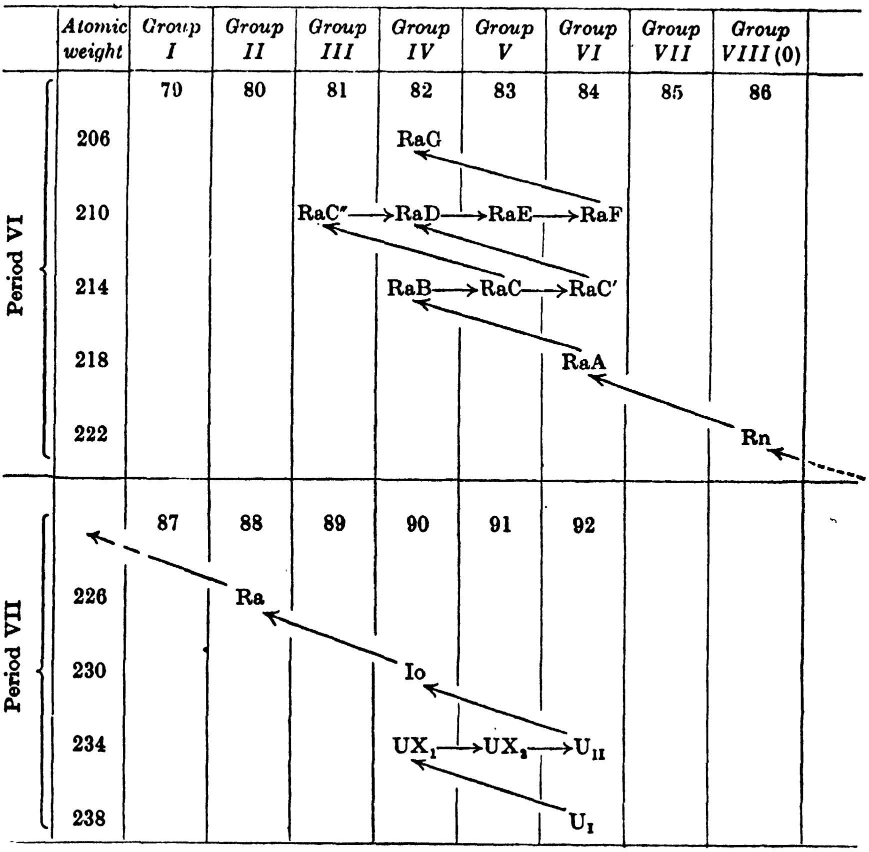
A particularly important relation is revealed when we apply this method of grouping the radio-elements to obtain the solution of the question as to what changes in the chemical nature of the elements ensue from the emission of α- or of β-particles. In other words, how does the chemical nature of the radio-elements change as we pass from one element to another within a transformation series ? We are enabled to recognize the required relation by comparing Tables 34 and 35 (pp. 143, 144) which give the sequence of and type of radiation emitted by the radio-elements, with Table 38, which shows the positions of the radio-elements in the periodic classification. Such a comparison leads to the following 'displacement law':

The emission of an α-particle gives rise to the formation of an element situated two places lower down in the natural sequence of the elements than the disintegrating element, whereas the emission of a β-particle leads to the formation of an element that is situated one place higher in the system.

In this consideration we regard the whole of the chemical elements as being arranged in a continuous series, in much the same way as they were represented by Lothar Meyer in his atomic volume curve (cf. Fig. 24, p. 64). In Mendeléeff's table we must remember that the individual periods are only portions of this continuous series, and that consequently the transition of an element in the zero group to a position situated two places lower down in the series signifies the appearance of an element in the sixth group of the preceding horizontal row. If this is clear, Table 39 will be immediately intelligible. It shows the

TABLE 39

Arrangement of the Uranium-Radium Series in the Periodic Classification



	Atomic weight	*Group I*	*Group II*	*Group III*	*Group IV*	*Group V*	*Group VI*	*Group VII*	*Group VIII (0)*
Period VI		79	80	81	82	83	84	85	86
	206				RaG				
	210			RaC″	RaD	RaE	RaF		
	214				RaB	RaC	RaC′		
	218						RaA		
	222								Rn
Period VII		87	88	89	90	91	92		
	226		Ra						
	230				Io				
	234				UX_1	UX_2	U_{II}		
	238						U_I		

uranium-radium series arranged in the last two horizontal rows of the periodic classification. In this table an arrow directed towards the left, and always embracing two groups, indicates the emission of an α-particle, whereas an arrow directed to the right, and reaching only the next group, signifies a β-transformation. Corresponding to the diminution in atomic weight, the arrows representing α-transformations are inclined to the horizontal. In particular, we notice that for RaC, which suffers 'dual' disintegration (see p. 142), the displacement law holds both for the product resulting from the emission of an α-particle and for that attendant upon a β-disintegration; i.e. RaC″ is situated two places lower and RaC′ one place higher than RaC. The validity of the displacement law for the actinium and for the thorium series can be represented diagrammatically in an analogous manner, but it can also be directly deduced from a comparison of Tables 34 and 38 or 35 and 38.†

From the displacement law it follows that when an α-transformation is succeeded by two β-transformations within one and the same disintegration series, the result must always be an isotope, as in the transition of U_I via UX_1 and UX_2 into U_{II}, which is isotopic with U_I. Two isotopes that are genetically related in this way must necessarily exhibit a difference in atomic weight of four units (= one α-particle), and from the consideration of such cases it is eminently clear that the atomic weight cannot be the determining factor in deciding the position of an element in the periodic classification. This conclusion is in splendid agreement with the views on the constitution of the atom already discussed in Chapter VIII. According to these, the position of an element in the periodic classification, or the 'atomic number' of an element, does not depend directly on its atomic weight, but primarily on the magnitude of the electrical charge of its atomic nucleus.

On the basis of this theory of atomic structure the radioactive displacement law follows as a matter of course. As a result of the emission of an α-particle, the charge on the nucleus diminishes by two positive units, and the newly formed element must therefore be situated two places lower in the periodic classification. A β-transformation, on the other hand, connotes an increase in the nuclear charge by one unit, since the production of a β-particle accompanies the transformation, in the nucleus, of a neutron into a proton (see Chap. IX, p. 94). From this it follows that the emission of a β-particle results in the formation of an element situated one place higher in the periodic classification. The successive loss of one α- and two β-particles must accordingly lead us back to the initial nuclear charge value. In other words, an isotope must be formed which is by four units lighter in atomic weight, as in the above-mentioned case of U_I and U_{II}.

It has been mentioned in earlier chapters (see p. 63 and p. 32) that a direct experimental evaluation of the nuclear charge for most of the elements has been possible from the investigation of X-ray spectra and of the scattering of the α-rays. From the viewpoint of the periodic classification the following results of these investigations were of especial interest.

(1) Isotopes like RaG and Pb have the same nuclear charge, and hence their insertion in one and the same place of the periodic classification is fully justified. Differences between them are based on the fact

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† Conversely, it has been mentioned (p. 146) that for the radio-elements of shortest life, the recognition of their chemical nature and their consequent inclusion in the table of isotopes were only rendered possible by the assumption of the validity of the displacement law.

that, although they have the same nuclear charge, the number or at least the arrangement of the structural components of the nucleus is different.

(2) Speaking generally, the chemical elements should be arranged in the sequence of their atomic weights; but it has long been known that this rule stands in contradiction to the chemical nature of the elements in three places in the periodic classification. For the three pairs of elements, argon-potassium, tellurium-iodine, and cobalt-nickel, the heavier element had to precede the lighter in the classification. This sequence of the elements in the periodic classification, established on the basis of their chemical behaviour, entirely corresponds to the succession of the nuclear charge values. Accordingly, if the value of the nuclear charge, instead of the atomic weight, be taken as the basis of the arrangement of the elements in the classification, the contradiction above referred to vanishes.

(3) In the interval between hydrogen (1) and uranium (92) the bearers of all the 92 nuclear charge values are known, with the exception of four. The missing elements have the atomic numbers:

43. eka-manganese;
61. rare earth;
85. eka-iodine;
87. eka-caesium.

These are the only chemical elements still unknown. Thus whereas there is possibly a large number of isotopes still to be discovered, the detection of new elements, apart from the four mentioned above, is only to be expected if elements exist with yet higher nuclear charge value than that of uranium (cf. p. 117 et seq.). The investigations described thus lead to the important result that we are able quite definitely to limit the expectations of chemistry, which had to proceed essentially in an inductive manner in its search for new elements, even after the formulation of the periodic classification.

From X-ray spectra we can, in the first place, fix only the atomic number of the missing elements. Here we may compare Fig. 24 (p. 64), which at once reveals that the regularity of the straight line would be immediately interrupted if, in the four stated positions of the abscissa, we were not to leave gaps for the elements yet unknown, but plot the values of the ordinates for the succeeding elements. On the other hand, we cannot gain information directly from the atomic number about the chemical nature of an element, because from amongst the four 'atomic analogues' of Mendeléef knowledge of the atomic number enlightens us only as to the 'horizontal neighbours', but not as to the 'group neighbours' of the element, and these are still more

TABLE 40

Periodic Classification of the Chemical Elements. (Long-period Mode of Presentation)

Period	*Group*																	
	1	*2*	*3*	*4*	*5*	*6*	*7*	*8*	*9*	*10*	*11*	*12*	*13*	*14*	*15*	*16*	*17*	*18*
I																	1 H 1·0078	2 He 4·002
II	3 Li 6·940	4 Be 9·02											5 B 10·82	6 C 12·01	7 N 14·008	8 O 16·0000	9 F 19·000	10 Ne 20·183
III	11 Na 22·994	12 Mg 24·32											13 Al 26·97	14 Si 28·06	15 P 30·978	16 S 32·06	17 Cl 35·457	18 Ar 39·944
IV	19 K 39·096	20 Ca 40·08	21 Sc 45·10	22 Ti 47·90	23 V 50·95	24 Cr 52·01	25 Mn 54·93	26 Fe 55·84	27 Co 58·94	28 Ni 58·69	29 Cu 63·57	30 Zn 65·38	31 Ga 69·72	32 Ge 72·60	33 As 74·91	34 Se 78·96	35 Br 79·916	36 Kr 83·7
V	37 Rb 85·48	38 Sr 87·63	39 Y 88·92	40 Zr 91·22	41 Nb 92·91	42 Mo 95·95	43 —	44 Ru 101·7	45 Rh 102·91	46 Pd 106·7	47 Ag 107·880	48 Cd 112·41	49 In 114·76	50 Sn 118·70	51 Sb 121·76	52 Te 127·61	53 I 126·92	54 X 131·3
VI	55 Cs 132·91	56 Ba 137·36	57–71 Rare Earths†	72 Hf 178·6	73 Ta 180·88	74 W 184·0	75 Re 186·31	76 Os 191·5	77 Ir 193·1	78 Pt 195·23	79 Au 197·2	80 Hg 200·61	81 Tl 204·39	82 Pb 207·21	83 Bi 209·00	84 Po 210	85 —	86 Rn 222
VII	87 —	88 Ra 226·05	89 Ac	90 Th 232·12	91 Pa	92 U 238·07												

† Rare Earths.

VI 57–71	57 La 138·92	58 Ce 140·13	59 Pr 140·92	60 Nd 144·27	61 —	62 Sm 150·43	63 Eu 152·0	64 Gd 156·9	65 Tb 159·2	66 Dy 162·46	67 Ho 163·5	68 Er 167·2	69 Tu 169·4	70 Yb 173·04	71 Lu 175·0

TABLE 41

Periodic Classification of the Chemical Elements. (Short-period Mode of Presentation)

Period	*Group I*		*Group II*		*Group III*		*Group IV*		*Group V*		*Group VI*		*Group VII*		*Group VIII*	
	a	*b*	*a*	*b*	*a*	*b*	*a*	*b*	*a*	*b*	*a*	*b*	*a*	*b*	*a*	*b*
I														1 H 1·0078		2 He 4·002
II	3 Li 6·940		4 Be 9·02			5 B 10·82		6 C 12·01		7 N 14·008		8 O 16·0000		9 F 19·000		10 Ne 20·183
III	11 Na 22·994		12 Mg 24·32			13 Al 26·97		14 Si 28·06		15 P 30·978		16 S 32·06		17 Cl 35·457		18 Ar 39·944
IV	19 K 39·096	29 Cu 63·57	20 Ca 40·08	30 Zn 65·38	21 Sc 45·10	31 Ga 69·72	22 Ti 47·90	32 Ge 72·60	23 V 50·95	33 As 74·91	24 Cr 52·01	34 Se 78·96	25 Mn 54·93	35 Br 79·916	26 Fe 55·84 27 Co 58·94 28 Ni 58·69	36 Kr 83·7
V	37 Rb 85·48	47 Ag 107·880	38 Sr 87·63	48 Cd 112·41	39 Y 88·92	49 In 114·76	40 Zr 91·22	50 Sn 118·70	41 Nb 92·91	51 Sb 121·76	42 Mo 95·95	52 Te 127·61	43 —	53 I 126·92	44 Ru 101·7 45 Rh 102·91 46 Pd 106·7	54 X 131·3
VI	55 Cs 132·91	79 Au 197·2	56 Ba 137·36	80 Hg 200·61	57 to 71 Rare Earths†	81 Tl 204·39	72 Hf 178·6	82 Pb 207·21	73 Ta 180·88	83 Bi 209·00	74 W 184·0	84 Po 210	75 Re 186·31	85 —	76 Os 191·5 77 Ir 193·1 78 Pt 195·23	86 Rn 222
VII	87 —		88 Ra 226·05		89 Ac		90 Th 232·12		91 Pa		92 U 238·07					

† Rare Earths.

VI 57–71	57 La 138·92	58 Ce 140·13	59 Pr 140·92	60 Nd 144·27	61 —	62 Sm 150·43	63 Eu 152·0	64 Gd 156·9	65 Tb 159·2	66 Dy 162·46	67 Ho 163·5	68 Er 167·2	69 Tu 169·4	70 Yb 173·04	71 Lu 175·0

important for the prediction of chemical properties. But since the position in the periodic classification is fixed by the atomic number, we can also recognize the chemical nature of the missing elements by making use of this classification. This we have already done above, by using the terminology 'eka' introduced by Mendeléef for the higher homologue.

Tables 40 and 41 represent the periodic classification of the elements on the basis of these results, Table 40 giving a 'long-period' and Table 41 a 'short-period' mode of presentation. Both forms have been given since each offers advantages for the consideration of certain regularities. Thus the long-period form is to be recommended in all considerations related to the theory of atomic structure (cf. Table 22 in Chapter VIII), whereas the short-period form epitomizes in a clear way particularly the laws of chemical valency.

It should be mentioned that the elements hafnium (72) and rhenium (75) were discovered only in recent years, and on the basis of the conclusions just described. At the time when the method of determining the atomic number with the aid of X-ray spectroscopy became known, it was necessary, in allotting atomic numbers to the chemical elements, to leave gaps not only in the above-mentioned four positions, but also in the positions 72 and 75. In the case of the element 75 (rhenium), the regularities in the periodic classification referred to above allowed definite predictions on the chemical nature of the missing substance to be made, so that the element could be looked for with success. The periodic classification in its old form allowed no decision to be arrived at as to whether the element 72 (hafnium) was one of the rare earth metals, or a homologue of zirconium. A reliable statement in the sense of the latter alternative was first rendered possible by the theory of atomic structure (cf. Table 22, p. 85). For both elements the first and at the same time the most certain proof that the missing substances were present in the mineral mixture under investigation resulted from observation of the X-ray spectrum. X-ray spectroscopy, accordingly, has proved its worth not only in revealing the gaps in the periodic classification of the elements, but also as a tool in analytical chemistry, and without it the successful search for elements yet undiscovered would have been rendered incomparably more difficult, if not quite impossible.

Information has already been given in Table 38 (p. 152) on the different forms in which the elements of the last two horizontal rows of the periodic classification occur, beginning with thallium. The isotopes of the inactive elements are shown in Table 43 (p. 182). (See Chapter XIX.)

XVII

THE CHEMICAL BEHAVIOUR OF EXTREMELY SMALL QUANTITIES OF A SUBSTANCE

FROM the genetic relation existing between the radio-elements it follows that we can obtain only extremely small quantities of any radio-elements of short life (see radioactive equilibrium, p. 131). Thus if we possess a quantity of radium as large as one gram, under the most favourable circumstances we can obtain from it about 13 mg. of the relatively long-lived radio-element RaD, whereas we can obtain only 0·22 mg. of polonium, and as little as 10^{-16} mg. of the very short-lived RaC′. For this reason the chemistry of the short-lived radio-elements is a chemistry of substances available in extremely small quantities, and therein lies its particular uniqueness.† From amongst the reactions especially characteristic of every chemical element, those of a spectral or other optical nature are generally inapplicable, as their sensitiveness is insufficient. We are restricted to the determination of the solubility of the elements and of their compounds, or to that of their electrochemical behaviour and the like, whereas the usual methods of chemistry always have to be supplemented by electroscopic measurements.

1. The Solubility and Velocity of Solution of Extremely Small Quantities

Owing to the smallness of the amounts available, we cannot prepare a saturated solution of the short-lived radio-elements, and hence the direct determination of the solubility is not possible. But we can determine the relative velocity of solution. Thus if we immerse the surface, say a piece of gold sheet, on which we have collected the active deposit of radium, into a dilute solution of an acid for only a few seconds, we find that a greater fraction of the atoms of RaB than of RaC have gone into solution, whereas the fraction of RaA atoms dissolved is still smaller. The electrochemical nature of these substances thus diminishes down the series RaA $\rightarrow$ RaC $\rightarrow$ RaB, in accordance with the chemical behaviour we should expect for the elements Po $\rightarrow$ Bi $\rightarrow$ Pb.

In individual cases we can also infer from the velocity of solution the existence of definite compounds of the radio-elements concerned. For example, from the much smaller velocity of solution of the ThB deposited on the anode, as compared with that on the cathode, we can infer the existence of a ThB-peroxide of difficult solubility on the anode.

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† The property of radioactivity influences the chemical behaviour only in the case of strong activities, and then in the same way as an external source would do (see Chap. XXIV, § 5).

This conclusion is confirmed by the observation that this difference in the velocities of solution can be annulled by the addition of a reducing agent to the acid used as solvent.

2. The Electrolytic Deposition of Small Quantities

The converse process to the solution of a metal is to be found in its electrolytic deposition. Thus if we electrolyse a solution of the active deposit of radium, RaA is deposited most readily, and RaB only with difficulty. If we perform the electrolysis with a small current, i.e. if we keep above a certain value of the cathode potential, it is possible to separate electrolytically the most noble of the available radio-elements in a very pure state. This method is of great importance especially in the preparation of polonium, which can be obtained in this way from solutions of the salts of 'radio-lead', as these contain an isotope of bismuth (RaE) and three isotopes of lead (RaD, RaG, Pb), in addition to Po. Further details are given on p. 214. Instead of by electrolysis, the separation of the radio-elements can also be effected by dipping a less noble metal into the solution of the radio-element concerned; this is analogous to the deposition of copper from a solution of a copper salt on a sheet of zinc. In radioactive investigations we very often require a preparation of RaC free from RaB. This can be obtained in the simplest manner by dipping a small sheet of nickel into an acid solution of the active deposit of radium which no longer contains the short-lived radio-element RaA.

Both the methods described lead to the establishment of a potential series of the radio-elements, the most noble member of which is polonium, and the least noble radium.

3. Determination of the Solubility of the Salts of Radio-elements by means of the Methods of Adsorption and Precipitation

The chemical behaviour of an element is characterized primarily by the solubility of its different compounds. In order to gain information about the chemical nature of the radio-elements it was therefore necessary to establish the solubility relations of their compounds. In the case of the short-lived elements, which are present only in quantities too small to be weighed, this was not possible by the study of their pure salts, but only by adsorption and co-precipitation methods, in combination with theoretical considerations.

The Method of Adsorption.

If we shake up a salt of low solubility, e.g. $BaSO_4$, with an aqueous solution of a salt the sulphate of which is readily soluble, e.g. bismuth sulphate, we find that an interchange takes place between the Ba-ions situated at the surface of the solid salt and the Bi-ions of the solution. Now the forces that strive to bind Bi- and SO_4-ions together to form a solid salt are much smaller than those operative in binding together Ba- and SO_4-ions. This follows from the much greater solubility of bismuth sulphate in dilute acids.† When the Bi-ions collide with the adsorbent they will only rarely be held by the SO_4-groups, and hence the adsorption will be small. The behaviour will be different, however, when the solution contains a salt such as $RaCl_2$, the cation of which forms with SO_4 a compound of low solubility. In this case the interchange between the Ra- and the Ba-ions at the crystal surface will be a very active one, and the radium will be bound or adsorbed by the surface of the $BaSO_4$. We can express this generally by saying:

A cation will be adsorbed by a salt of low solubility when it forms with the anion of the adsorbing salt a compound, the solubility of which in the solvent is small. The less the solubilities of the relevant compound and the adsorbent, the stronger will be the adsorption.

By means of this method it can be readily proved, for instance, that ThX and AcX (both types of Ra) form sulphates of very low solubility, and that the 'B'-products (types of lead) give rise to sulphates of fairly low solubility; on the other hand, the sulphates of the 'C'-products (types of Bi) and also of polonium are readily soluble. If the experiments are carried out in potassium hydroxide, in which the solubility of lead sulphate is known to be greater than that of bismuth sulphate, a correspondingly weaker adsorption of the B-products is found than of the C-products.

The Method of Precipitation.

Instead of establishing which salt of low solubility adsorbs the radio-element concerned, we may also proceed along the following closely related course. A soluble barium salt is added to the solution of the radio-elements, say the active deposit of thorium, and the Ba precipitated as sulphate. Whereas the ThB is also precipitated, the ThC remains in solution, from which we may likewise infer that the first compound (lead sulphate) is only soluble with difficulty, whilst the latter compound (bismuth sulphate) is readily soluble. *A radio-element will be precipitated in greater measure with a deposit of low solubility, the less soluble is its compound with the negative constituent (anion) of the precipitate.*

From the results of experiments on adsorption and precipitation it seems to follow that for the extremely small quantities involved in the case of the radio-elements, the essential condition for their inclusion

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† More accurately expressed, it is not solely a question of the magnitude of the forces holding the ions together in the crystal lattice, but of the excess of these forces over those which seek to bind the ions to molecules of water, or to hydrate the ions.

in the precipitate is primarily their low solubility. But more recent investigations have shown that the size and electric charge of the surface of the adsorbent are also weighty factors in adsorption. Moreover, in problems of co-precipitation, an important part seems to be played by the question as to whether the compound of the radio-element and the anion of the precipitate is isomorphous with the crystals of the precipitate, so that it can form mixed crystals with them. The relations are here so complicated that it is not yet possible to formulate rules embodying all processes in a unified scheme.

4. Volatility of Extremely Small Quantities

The observation of the proportion in which the different components of the active deposit volatilize when heated for a short time at high temperature enables us to form a conclusion as to the sequence of the boiling-points of the radio-elements concerned, or of their compounds. Thus in air at 700° C. none of the 'C'-products volatilize, whereas the 'B'-products are already appreciably volatile; from this it follows that ThB-oxide is more volatile than the oxide of ThC (lead and bismuth oxides respectively). This method can also be applied for the partial separation of radio-elements of short life.

Nevertheless, an accurate determination of the boiling-point of extremely small quantities is not possible, chiefly owing to the preponderance of the process of sublimation. Thus a quantity of radon of the order of 1 curie $= 0{\cdot}65$ mm.^{3} manifests a sharply defined boiling-point at $-62°$ C. and a freezing-point at $-71°$ C., but quantities of radon of the order of 10^{-7} curie no longer possess a definite boiling-point. Such small quantities already begin to sublime appreciably at $-164°$ C., and at $-125°$ C. practically no condensation at all can be recognized.

5. Velocity of Diffusion. Determination of Valency and the Degree of Dispersion. Colloidal Radio-elements

The determination of the velocity of diffusion in water of the ions of all the radio-elements with a half-value period not very much less than one hour can be performed by the usual method. In fact, the extremely small concentration is of particular advantage here, seeing that we have to operate with solutions as dilute as possible.

We can likewise determine with little difficulty the electrolytic mobility, or the velocity with which the radioactive ions move in an electric field of unit gradient. The ratio of this to the velocity of diffusion immediately reveals to us the charge carried by the ion concerned, and hence its valency. Since the mobilities of the majority of inorganic ions differ among themselves by only a few per cent., we can also evaluate the valency, which can only amount to a small integral number (1 to 4), directly from the velocity of diffusion. From the constants of diffusion 0·33, 0·46, and 0·66 cm.^{2} day^{-1} for radioactinium (thorium isotope), actinium (homologue of lanthanum), and actinium X (isotope of radium) respectively, we can for instance calculate that the respective valencies are 4, 3, and 2.

Diffusion experiments further show that under certain circumstances radio-elements can exist in the colloidal form. Thus in neutral and in weakly acid solution polonium is partially colloidal; it diffuses slowly, does not dialyse through a parchment membrane, and so on. This behaviour serves as the basis of a method of concentrating the polonium in radio-lead; the polonium remains behind in the dialyser, and the crystalloidal lead diffuses outwards through the parchment.

Isotopes of bismuth, too, show such a marked tendency to appear in colloidal form that when we filter a neutral or weakly acid solution of ThB+ThC, the latter (a type of bismuth) is for the most part retained by the filter.

A particularly striking confirmation of the colloidal nature of the solution of some of the radio-elements has been furnished by the following observation. When ions or other colloids of opposite sign are added to the solutions, the phenomena of the inversion of charge and precipitation characteristic of colloids are obtained in exactly the manner to be expected in such cases. Moreover, it has been proved by means of the photographic plate that radioactive colloidal particles have an invariable tendency to aggregation and sedimentation.

The molecular weight of the emanations (radon, thoron, actinon) can be approximately determined by observation of the velocity with which they flow through small orifices, and in this way, for actinon, values lying between 222 and 218 have been obtained.

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XVIII

APPLICATION OF THE RADIO-ELEMENTS AS INDICATORS IN PHYSICAL, CHEMICAL, AND BIOLOGICAL INVESTIGATIONS

As we have seen, many difficulties had to be overcome before the chemical nature of the natural radio-elements could be recognized. But now that we know the nature of all of them with great certainty, we can apply them conversely to obtain information on the behaviour of known substances in such small concentrations that they cannot be investigated by ordinary methods. Elements such as lead and bismuth, which exist both in the form of a stable type and as radioactive isotopes, are particularly amenable to investigations of this kind, for here it is possible, by mixing in varying proportions the stable element with a radioactive isotope, to bridge over the whole range from quantities that can only be detected by radioactive means to quantities that can be weighed. In this manner we are able to investigate completely the behaviour of one and the same chemical element over a very wide range. In problems of this kind, in which the radio-element is not the object but the agent of the investigation, we say that the radio-elements serve as 'indicators'.

In the choice of a radio-element suitable as an indicator it is advisable in general to find one that can be detected in quantities as small as possible. We must bear in mind, however, that the half-value period (T) varies in inverse proportion to the strength of the radiation (see Chap. XII), and that an element that is measurable in small amounts necessarily possesses a short half-value period. For this reason it would never be possible to make use of say RaC′ ($T = 10^{-6}$ sec.) or the more strongly active ThC′ ($T = 10^{-11}$ sec.) as indicators. Moreover, the only three indicators available for the element thallium are already too short-lived for many experiments.

It follows that for our present purposes we cannot draw upon all the radio-elements. Only those included in Table 42 have any practical application, and the ones most frequently to be recommended are printed in italics.

Radio-elements like ionium, uranium X_1, and uranium X_2, which have no inactive isotopes, are also frequently of service, for with their aid we can raise the intensity of the radiation of their less virile isotopes which, although themselves radioactive, have a longer life and are therefore less active and more difficult to measure. Thus uranium X_1 is mentioned in the table as an indicator capable of being used for thorium. In the same way we can apply thorium X as an indicator

TABLE 42

Radioactive Indicators

Atomic number	*Element*	*Indicator*
81	Thallium	Radium C$''$ Thorium C$''$ *Actinium C$''$*
82	Lead	Radium B *Radium D* *Thorium B* Actinium B
83	Bismuth	Radium C *Radium E* *Thorium C* Actinium C
84	Polonium	Radium A
86	Radon	*Thoron* Actinon
88	Radium	*Thorium X* Actinium X
89	Actinium	Mesothorium 2
90	Thorium	Ionium Radiothorium Radioactinium *Uranium X_1*
91	Proto-actinium	Uranium X_2

for radium, and radium A for polonium. But radium and polonium themselves can also be used as 'indicators' in some investigations; for if in the narrower sense we understand the application of the radio-elements as 'indicators' to mean that it only refers to experiments in which a radio-element takes the place of its active or inactive isotope, then in the broader sense this expression can also be used for all those experiments in which a radio-element serves as an aid in the examination of the behaviour of matter present in the smallest amounts.

In addition to the natural radio-elements hitherto considered, the recently discovered and artificially produced radio-elements can also be used as indicators. As explained more fully in Chapter X, the bombardment of certain of the elements by α-particles results in the production of radioactive substances which emit positrons when they disintegrate; and in the bombardment of almost all the elements by neutrons new types of atoms are formed, which are transformed by the emission of electrons. Provided that these artificial radio-elements are produced in sufficient quantity to be detectable with ease, and that they have a period of life reckoned in minutes or more, they are straightway adapted also to be used as radioactive indicators. Although the discovery of the artificial radio-elements is of quite recent date, there are already available a significant number of investigations on these lines, and there can be no doubt that in the future the artificial radio-elements will surpass the natural radio-elements in their importance as indicators; for in the study of practically all the elements in the periodic classification they place at our disposal the corresponding active isotopes, including those of carbon, phosphorus, sodium, calcium, iron, etc., which are of especial importance in biological experiments. Sometimes it is even possible to produce the radioactivity directly in the chemical compound of the element to be investigated [1].

It may be mentioned that in those rare cases in which inactive isotopes are readily accessible they can be used as indicators in fundamentally the same manner, though other methods of measurement must be used. Thus instead of making electrometric measurements it is necessary to determine the density or to make determinations of the thermal conductivity, and so forth. Such experiments have been carried out with deuterium ^{2}D and the oxygen and nitrogen isotopes ^{17}O, and ^{15}N but further discussion of them does not belong here.

The applications of 'radio-elements as indicators' fall mainly into three groups; with their aid:

(A) We can study the behaviour of minute quantities of a radio-element, or of an indicated element,† down to invisible and unweighable amounts.

(B) We can determine very small fractions of an indicated substance which has been separated from the main part by solution, diffusion, etc.

(C) By mixing only a fraction of a stable element with its radioactive isotope we can differentiate this fraction from the rest of the element, and then *study the exchange of atoms of identical chemical properties*. It is especially this use of indicated atoms which opens the door to otherwise insoluble problems.

A. THE STUDY OF MINUTE QUANTITIES OF AN ELEMENT

A problem frequently met with in analytical chemistry is the distribution of traces of an element between precipitate and solution; minute amounts of many elements are included either by co-precipitation or by adsorption on the crystals of a precipitate or, conversely, remain dissolved when the bulk of the element is removed. In the case of radio-elements or indicated elements such an imponderable quantity can be

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† An *indicated element* is an element containing a small amount of an isotopic element, which serves as an indicator for the purpose of detection or measurement.

determined without the slightest difficulty. Studies of this type have shown that methods of chemical separation, which are reliable for normal weights of matter handled in an analytical laboratory, fail in the collection of small amounts.

For example, if we want to determine the lead content of ordinary rocks or meteorites, which is only of the order of a thousandth of one per cent., we find its quantitative separation practically impossible; in this case the application of radium D as an indicator has proved to be very useful. For this purpose a known amount of RaD was added to the solution of the material, and by electrolysis this was deposited anodically as RaD O_2 together with the contained lead. Thus, if in this process only 80 per cent. of the amount of RaD originally added were regained by electrolysis, it follows that the yield of the lead analysis was likewise only to the extent of 80 per cent., so that the mass of the deposit must be increased by 25 per cent. in order to obtain a quantitative value for the lead content of the rock.

In this simple example the radio-element RaD serves directly as an indicator of the element to be estimated, i.e. lead. But the range of application of the radioactive method in analytical chemistry can be extended considerably if the element lead, activated by the radio-element ThB, is in its turn only the reagent for the substance (e.g. ammonia) in which we are interested. This method of 'Radiometric Microanalysis' has rendered it possible, for instance, to carry out a determination of the nitrogen content of organic substances to the order of magnitude of some ten-thousandths of a milligram. The decomposition of the organic substance with sulphuric acid and the distillation from caustic soda of the ammonia formed is essentially the same as in the well-known micro-Kjeldahl process, except that several special precautions have to be taken, corresponding to the higher sensitivity aimed at. The ammonia distilled over is collected in a standardized (but not radioactively activated) solution of lead nitrate; the precipitated lead hydroxide is centrifuged off, and then, in aliquot parts of the solution, the lead that has not been precipitated is determined with the help of a radioactive indicator method. This is done by mixing each 1 c.c. of the solution, freed from the precipitate by centrifuging, with 0·5 c.c. of a radioactivated N/1,000 solution of lead nitrate, and then precipitating with 0·5 c.c. of a N/1,000 solution of potassium chromate. It follows from the fundamental facts of isotopy that the indicator substance ThB that has been added with the 0·5 c.c. of the lead nitrate solution will be distributed between the precipitate and the solution in accordance with the available amount of total lead. Hence the more lead there is present in the lead solution under estimation, i.e. the less ammonia there has been introduced into it, the greater will be the activity remaining in the solution. By distilling over known amounts of ammonia and maintaining always the same conditions of experiment, the electroscope can be calibrated directly in terms of ammonia.

B. THE DETERMINATION OF VERY SMALL FRACTIONS OF AN INDICATED SUBSTANCE

The solubility of lead chromate at room temperature is too small to be determined with accuracy by gravimetric methods. It is of course possible to fall back on conductivity measurements or to calculate the solubility product from equilibrium constants; but for concentrations so small as that of a saturated solution of lead chromate both of these methods are tedious and subject to several sources of error. On the other hand, with the help of a radioactive indicator we can determine the solubility of lead chromate just as readily as we can perform a simple weighing operation. For this purpose we add a definite quantity of ThB to a known amount of a soluble lead salt. The radioactivity of this indicated substance can be measured in terms of any arbitrary unit such as the number of scale divisions per minute obtained with the electroscope used. Thus if 10,000 such units of ThB have been thoroughly mixed with 10 mg. of lead, it is clear that one unit of ThB will then always necessarily indicate the presence of 0·001 mg. lead. Now we can prepare lead chromate from the artificially activated lead by the usual chemical methods. If we have kept the saturated solution of this compound sufficiently long in a thermostat at the desired temperature, we can determine its concentration by evaporating to dryness a few c.c. of the solution, and measuring with the electroscope the activity of the almost invisible residue. A special advantage of the method is that the presence of foreign ions interferes in no way with the measurements, for of course only the radioactive substance influences the electroscope.

Another problem in which a very small quantity of the initial amount of an element had to be detected was encountered in the search for the hydride of bismuth, the analogue of arsine and stibine. As a result of many fruitless attempts to prepare it, the conclusion was reached that, if it could be prepared at all, this compound would only be obtainable in amounts corresponding to vanishingly small fractions of the initial quantities of material used, and that it would thus readily escape detection by the usual chemical methods. But the sensitivity of radioactive measurements can be graded down so far as to encompass both qualitatively and quantitatively even the ten-millionth part of the initial amount of the substance. This enormous extension of the range of measurement is necessary if we are to study the condensation and revolatilization of bismuth hydride; but the fact of its existence can be shown by relatively simple means, and even as a lecture experiment. When magnesium filings are coated with the bismuth isotopes ThC or RaC and dissolved in dilute hydrochloric acid, the volatile hydrides of these radio-elements are evolved, and can be condensed at the temperature of liquid air, or decomposed in a hot glass tube and measured. By means of the radioactive method just described it was possible to accumulate experience as to the best mode of preparation and on the degree of stability of bismuth hydride, and it was but a short step to the long-aspired goal of preparing this compound also from inactive bismuth by similarly dissolving a magnesium-bismuth alloy in hydrochloric acid. It may be mentioned that the quantity of bismuth with which the existence of this volatile compound was first recognized amounted to only 10^{-15} gm., and that it has been found possible with the help of radioactive indicators to detect the existence also of gaseous lead hydride, the yield of which is still smaller.

Technical investigations can also benefit from the application of radioactive indicators. It has been found possible to determine with speed and accuracy and quantitatively the different permeabilities to air of almost completely air-tight gummed materials (gas-mask fabrics) by using air containing traces of radon. For precipitates in which radium is imbedded, an alteration in their surfaces can be recognized simply and with certainty from the change in their emanating power in respect of radon. This method has been shown to be one of much promise in the solution of practical problems, such as the examination of the condition of the surface of glass.

In the treatment of syphilis, by making use of radium E as an indicator, the retention of bismuth in the organism was investigated in detail. For a long time after treatment a considerable amount of the metal remains in the body, maintaining an anti-syphilitic effect; the important matter was to gain information as to the rate at which different bismuth compounds are resorbed after injection. A further investigation concerned the retention of lead (indicator: ThB or RaD) and of thorium (indicator: UX_1 or Io) both in human and animal bodies; extended to the behaviour of tumours, it proved that cancerous tissues retained appreciably larger amounts of bismuth than the corresponding healthy tissue.

While in the examples quoted the problem was to determine fractions of the original substance, an experiment may be mentioned in which the total amount of a substance is unknown, but can be inferred from an activity measurement of a fractional part of it. The flow of water in a river can be determined by adding a small quantity of a radium salt as an indicator [2]. For this purpose it is only necessary to allow the radioactive solution to flow for a given time at constant rate into the river, and then to take a sample of the river water sufficiently far downstream to ensure that the radium solution has been thoroughly and uniformly mixed with the water of the river. A simple measurement of the activity of this sample immediately reveals the quantity of river water with which the added radioactive solution has been diluted, and thus the flow of the river. If, instead of the radioactive solution, a solution of an inactive salt is used, a method that has occasionally been applied, the performance of the experiment is much more complicated and costly, because of the large quantities of the salt that must be used in order that it may be detectable at such high dilutions.

In a similar manner, attempts have been made to determine the quantity of blood in living animals, but in comparison these experiments are much more prone to the objection that part of the radioactive substance may be lost by adsorption and chemical exchange.

C. THE EXCHANGE OF ATOMS OF IDENTICAL CHEMICAL PROPERTIES

The interchange of atoms and molecules within a homogeneous substance can be followed *only* by means of isotopic indicators. The simplest example of such a kinetic problem is presented by the addition of an activated solution of $Pb(NO_3)_2$ to an inactive solution of $PbCl_2$ of equivalent strength. By crystallizing from this mixture a sample of lead chloride, we can determine whether it contains not only lead atoms which were linked with chlorine before the mixing, but also lead atoms originally combined in the form of nitrate. As we should expect, according to the theory of electrolytic dissociation, the result of such a determination demonstrates that the lead atoms in the crystallized lead chloride are derived in equal numbers from the two initial solutions. On the other hand, let us say, if activated lead nitrate and inactive tetraphenyl-lead, or another compound in which the lead atom is linked with carbon in a non-ionizable form, be simultaneously dissolved and then separated by crystallization, we are unable to detect any interchange of the lead atoms between the lead nitrate and the organic compound of lead.

In an analogous manner it has been possible to show that the charges of di-valent and tetra-valent lead ions are interchanged with the greatest of ease. After the discovery of artificial radioactivity it was also feasible to establish the same phenomenon with manganese ions of different valency, and to examine also the firmness of binding of iodine, bromine, etc., in organic compounds, as well as to study the mechanism of important reactions in organic chemistry. For instance, in the case of a molecule containing iodine, it was possible, by the use of iodine activated artificially, to give an affirmative answer to the question as to whether the racemization of an optically active compound is preceded by a substitution [3].

The interchange between the solid and liquid phases, as between metal and ion, or between a salt and its saturated solution, is more difficult to picture, and for this reason even more important to investigate than the positional interchange in solutions. These processes, as functions of time, can be readily followed by means of radioactive indicators; and such investigations can be made the basis of a method of measuring the surface area of powders, which is of importance in experiments on adsorption. This method is based on the fact that when a radio-element is added to a saturated solution of the isotopic adsorbent, in the form of powder, it must distribute itself uniformly in a very short time between the molecules of the dissolved part of the adsorbent and the molecules of the outermost layer of the undissolved part of the powder. From the diminution of the radioactivity of the solution in this way we can easily calculate the dimensions of the surface of the powder. The surface areas of lead sulphate, lead sulphide, bismuth phosphate, and of many other compounds have been measured in such a manner, and with the aid of the data so obtained it has been possible to prove that, for all these adsorbents, the limit of adsorption is reached before the adsorbed substance has formed a coherent layer even one molecule thick on the surface of the adsorbent.

Closely related is the method which made possible, by the use of radio-elements, an experimental investigation of the process of 'self-diffusion'. Here we are concerned with the fact predicted by the kinetic theory of matter that the atoms (or molecules) of a homogeneous gas or of a liquid move amongst themselves according to the same laws as govern the diffusion of one element into another. But since in the process of self-diffusion the objective properties of the element investigated remain entirely unchanged, this process could previously be carried out only as a 'conceptual experiment'. Not until the introduction of radioactive indicators was it possible to open up to observational study the phenomenon of self-diffusion. In performing the experiment, a lead cylinder was fused into a hard glass tube, three-quarters of the length of the cylinder consisting of ordinary lead and the remaining quarter of activated lead. For experiments of short duration ThB was used as indicator, whereas for those of long duration RaD was chosen. By carefully melting the lead in the tube and maintaining it at liquefying temperature for several days the indicated active atoms of lead were intermingled with the other lead atoms by diffusion; it was possible after solidification to investigate the distribution of the activity established in the column of lead during the experiment, and the constant of self-diffusion in molten lead could be calculated by the usual methods. By application of specially sensitive methods it was also possible to measure the coefficient of self-diffusion in solid lead. When RaD atoms are deposited on the surface of a thin sheet of lead they gradually diffuse into it, and this process can be followed by observation of the diminution in the number of scintillations or of the ionization due to α-rays, emitted by the succeeding product polonium.

In a similar way we can follow the change in position of the lead ions in heated crystalline lead chloride, and thence draw conclusions about the processes of diffusion and conduction of electricity in crystals.

After the discovery of artificial radioactivity it was possible to examine the circulation of phosphorus in human or animal systems [4]. The active form of phosphorus was produced from sulphur, and the atoms of phosphorus in food could be indicated by the addition to them of sodium phosphate of known activity. In this manner it was established, for instance, that in the course of 5 days, 22 per cent. of the phosphorus atoms in a man's food are evacuated by the kidneys and 16 per cent. by the bowels, whereas when the phosphorus atoms are introduced directly into the blood-stream the corresponding proportions eliminated are respectively 20·5 per cent. and 2·5 per cent. in the same time. The remainder of the phosphorus atoms enter into the structure of various organs, and above all into the bones, whence they are sooner or later displaced by the arrival of fresh phosphorus atoms. It was possible to measure the rates of formation of various organic phosphorus compounds (viz. phosphorus esters, lecithin, etc.) present in the blood and in the different organs, as well as to determine the distribution of the phosphorus atoms taken up into the system between individual bones, muscles, liver, kidneys, teeth, and so on. The study of the behaviour of the phosphorus in the brain was of special interest. If some ten hours after administration of radioactive sodium phosphate to rats or other animals the phosphatides of the brain are extracted, they are found to contain radioactive phosphorus atoms. Now we know that phosphorus atoms present in a phosphatide molecule take no part in simple exchange processes; therefore the radioactive phosphorus atoms can only have entered these molecules during their synthesis. Hence, from the presence of radioactive phosphatides in the brain tissue of fully grown animals after the intake of radioactive sodium phosphate, we can conclude that these tissues, contrary to general assumption, are constantly regenerated; presumably the lecithin molecule, under enzyme action, is broken down and rebuilt.

Analogous experiments undertaken with maize and sunflowers showed that most of the phosphorus atoms are found in the leaves, and that they are mobile. Investigation revealed that an individual phosphorus atom which is to-day situated in one leaf in the course of a few days will very probably be found in another leaf, while in seeds no exchange takes place between the phosphorus atoms of the germ and of the endosperm [5].

In conclusion it may be emphasized that in the measurement of the rays we are not confined to electrical methods, for sometimes the observation of scintillations or photographic methods are to be preferred. We have already given one example of the application of the scintillation screen in connexion with the measurement of the coefficient of self-diffusion in solid lead. The photographic plate may offer special advantages over the electroscope in so far as with it the particular spot at which a chemical change has taken place can be rendered visible. When an investigation was made, in experiments previously described, into the kinetic interchange of molecules between the crystals of a solid substance and an activated saturated solution of the substance, it was found that for certain crystals the number of molecules taking part in the exchange was distinctly smaller than the number of molecules comprising the outermost layer of the crystals. This proved that the kinetic exchange takes place only at privileged positions in the surface. The actual positions could not of course be found by means of the electroscope used in this investigation, because only the total effect of all the rays is measured by the electroscope. But when the crystals were allowed to act on a photographic plate, it was immediately revealed that the radioactive atoms were situated almost wholly at the edges of the crystals, so that the exchange between the crystals and the radioactive solution had taken place at these edges. In this way the seat of reactions can be recognized to a nicety.

The examples we have given should suffice to show that the method of radioactive indicators is capable of multifarious applications. It is to be hoped that in the future an increasing number of workers will make use of these indicators and in this way facilitate the solution of many problems.

In particular, it is perhaps not superfluous to emphasize that such experiments involve by no means heavy costs. As will be seen from the above examples, many of the experiments have been carried out with thorium B or thorium C as indicators. Both of these substances are themselves quite short-lived ($T_{\mathrm{ThB}} = 10{\cdot}6$ hours; $T_{\mathrm{ThC}} = 60{\cdot}5$ minutes) and the quantities of radioactive substance used in the individual experiments have therefore practically no monetary value, for these amounts are freshly obtained before each experiment from the parent substance radiothorium (see p. 216 et seq.), which itself remains completely unchanged and only decays with a half-value period $T = 1{\cdot}90$ years. Since '1 mg.' radiothorium (i.e. a quantity whose γ-radiation is equivalent to that of 1 mg. radium) supplies an adequate amount of ThB and ThC for almost all indicator experiments, and this quantity is always for the most part regenerated 24 hours after removal of the preparation from the radiothorium, it is possible with '1 mg.' radiothorium to perform an indicator experiment daily either with ThB or with ThC throughout a period of several years. At the present time '1 mg.' radiothorium costs about eight pounds. In many experiments distinctly smaller amounts of radiothorium will suffice; on the other hand of course there are also experiments in which greater activities are desirable, just as occasionally it is also impossible to avoid using other indicators like RaD and RaE, which are more difficult to obtain.

The acquisition also of artificial radio-elements as indicators does not necessarily involve large monetary expenditure, especially when, as is often the case, radon can be obtained from hospitals and similar institutions. A small glass tube filled as full as possible with very finely grained beryllium powder and with radon constitutes a source of neutrons which can be used for several days for the production of artificial radio-elements. For many experiments quantities of about 50 millicuries of radon suffice; for others, however, hundreds of millicuries of radon are necessary, and still stronger active preparations can be obtained by bombardment with deuterons that have been accelerated say in a cyclotron (see p. 290). When a source of neutrons is required for continuous use it is of course desirable to mix a salt of radium instead of radon with beryllium powder. The costs can frequently be reduced by renting radium for a specified period instead of buying it.

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XIX

ISOTOPY OF ELEMENTS WHICH ARE NOT RADIOACTIVE

In Chapter XV a description was given of how the phenomenon of isotopy was discovered for the radioactive elements. Isotopes are substances that differ from each other in respect of their radioactive properties and atomic weights, but in general they cannot be separated from each other by chemical means. From this it was necessary to draw the conclusion that the inability to resolve an element chemically does not constitute proof of the law formulated by Dalton, according to which the atoms of one and the same element all have the same weight. It was a curious coincidence in point of time that the factual incorrectness of Dalton's assumption was proved simultaneously by other investigations. Although these researches, which establish the existence of isotopes in the field of the non-radioactive elements, are only loosely connected with the theme of this book, we shall briefly discuss the principle of the methods used and the results obtained, because they are of great importance in a consideration of the position of the radioactive atoms within the entire system of the elements.

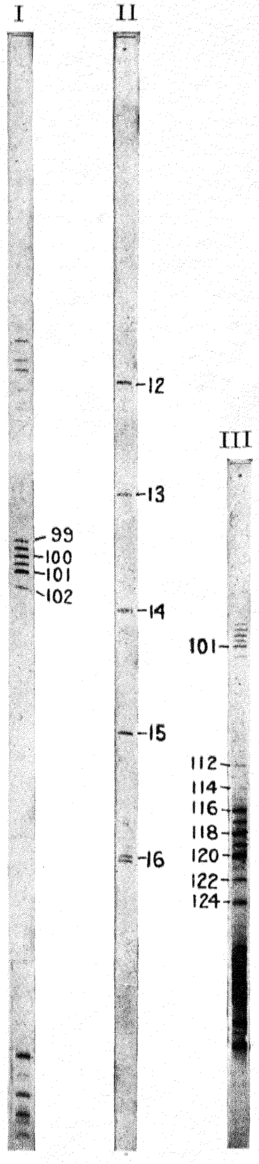
The first indication that an element can contain atoms of different weight was found in the study of positive rays for the case of neon. The systematic development of these investigations led to the construction of the 'mass spectrograph'. For those elements that consist of a mixture of different types of atoms, this apparatus enables us to establish individually the masses of these atomic types, whereas it is well known that the chemical methods for the determination of the 'atomic weight' only yield mean values in the case of mixed elements. For this reason it is better to use the term 'combining weight' instead of 'atomic weight' (see Chap. XXI).

The following principle lies at the basis of the mass spectrograph: The element to be investigated is subjected to electrical discharge in a highly evacuated tube, whereby its molecules and atoms in part assume one or more positive charges, and as positive rays they travel towards the cathode and pass through a slit in this into the experimental chamber. Here the particles are first deflected by an electrostatic field and then they pass through a magnetic field so arranged that deflexion now takes place in the opposite direction, and they finally strike a photographic plate. By suitable choice of the strengths of the electric and magnetic fields, and by the use of suitable diaphragms and an appropriate position of the photographic plate, it can be ensured that all the particles with a constant value of m/e are brought to the same place on the photographic plate, even when their velocities fluctuate within a certain range. When particles of the same mass *m* bear a double electric charge, they are deflected to another position on the photographic plate, in fact, to the same place as all particles of mass $\frac{1}{2}m$ and single charge *e* would strike. In this connexion the terminology of optical spectroscopy is applied, and the mass spectra so obtained are designated as spectra of the first, second, and higher orders. The value of m/e corresponding to the individual lines is found by comparison with the neighbouring lines of known atoms and molecules such as C, CO, CO_2, O_2. The sensitivity of the method is so great that in the majority of cases an isotopic mixture in the proportion of 999:1 can still be recognized as a mixture of two types of atoms.

The photogram represented in Fig. 45 (Plate IV), which is taken from the publications of Aston, gives an idea of the sharpness with which these mass spectra can be obtained. For precisional determinations, somewhat different methods developed by Dempster and by Bainbridge are also found to be particularly suitable. The former is based on the fact that the charged atoms all traverse the same potential difference and so acquire the same energy, so that when they pass through a magnetic field, particles of equal mass and having the same velocity are brought into the collecting electrometric system without any special focusing device. If particles of different mass are present, and the magnetic field is held constant, they can be collected in turn according to their masses by appropriate adjustment of the accelerating potential, the required value of which is inversely proportional to the mass.

The 'whole number rule' was the most important result brought to light even by the earliest mass-spectroscopic investigations, and it has been consistently confirmed by the results of later experiments. By this we mean that the atomic weights of all types of atoms approximate very closely to whole numbers, when we assume the atomic weight of ^{16}O to be equal to 16·0000. Thus the large deviation in the atomic weight of chlorine (35·457) from an integral value is due to the fact that chlorine consists of two isotopic types of atom in the proportion of 76 per cent. of ^{35}Cl to 24 per cent. of ^{37}Cl. A marked deviation of the combining weight of an element from an integral value is thus in itself proof of the fact that we are dealing with a mixed element. Conversely, however, when the combining weight of an element is almost integral in value, we are not necessarily justified in concluding that we are dealing with a pure element. Bromine is an example of this, for it contains almost equal numbers of atoms of mass 79 and 81, so that its combining weight lies very close to the value 80.

That integral numbers occupy a privileged position in the matter of atomic weights can be readily understood from current ideas on the



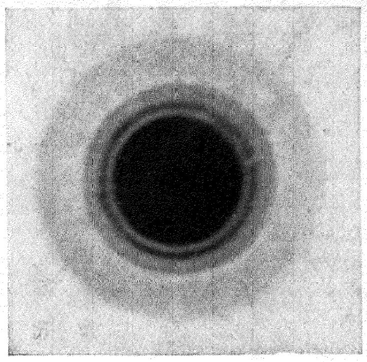


FIG. 52 (p. 270)
Normal Pleochroic Halo in Biotite
(Magnification *c.* 520)

FIG. 45 (p. 178). Mass Spectra (I shows the second order mass spectrum of Hg; II shows the lines of C, CH, CH$_2$, CH$_3$, CH$_4$, and that of ^{16}O directly alongside the last of these; III shows the 11 isotopes of tin, as well as xenon lines).

structure of atomic nuclei, since they are assumed to consist of only protons and neutrons (see Chap. IX). For practical as well as for theoretical reasons it is therefore advantageous to introduce the concept of 'mass-number' in addition to those of combining weight and atomic weight. From the practical point of view the mass-number is that whole number which lies nearest to the atomic weight; and in the theory of atomic structure it denotes the numerical sum of the protons and neutrons in the nucleus of the atom.

It is to be expected on theoretical grounds that the whole number rule will hold only as a first approximation, and deviations from it are of great importance in relation to the nuclear structure of the atom. Since in all theoretical considerations we are less concerned with the absolute than with the relative magnitude of the deviations from integral values, the idea of the 'packing fraction' has been introduced, by which we understand the difference between the mass of the atom and the integral part of its mass-number, divided by the mass-number. The packing fraction of ^{16}O is of course zero, since the masses of the atoms are referred to $^{16}O = 16{\cdot}0000$. Elements lighter than oxygen have positive values of the packing fraction; thus ^{1}H has a packing fraction 0·0081, and ^{4}He the value 0·00098. The atomic masses 20 to 200 (neon to mercury) have negative packing fractions; thus Sn has the value $-0{\cdot}00073$. The heaviest elements again have positive values of the packing fraction, as with thallium $+0{\cdot}00018$. As has been discussed in greater detail in Chapter X, the stability of atomic nuclei and the possibility of their mutual transmutation are related most intimately with these small differences in mass, so that one of the most important problems in mass-spectroscopy is the evaluation of them with the greatest possible accuracy.

None of the methods of mass spectroscopy is sufficiently sensitive to detect an isotopic atomic constituent when its proportion is considerably less than 0·001 of that of the isotope most strongly represented. As regards the existence of atomic types that are present in an element in such relatively small quantities, important information has been obtained by the examination of optical spectra.

In line spectra the differences due to the different masses of the isotopes generally lie beyond the limit of measurement. A very important exception to this is found in the case of hydrogen, whose heavier isotope ^{2}D has been recognized by a displacement of the lines of the Balmer series. By using spectrographs of very high resolving power, however, and studying the fine structure of line spectra, it has been possible with other elements also to obtain information on the existence and relative abundance of their isotopes. The isotope of lead 204 [1], five isotopes of platinum [2], and an isotope of lutecium [3] have been detected by this method, although the abundance of the last-named amounts to only 1·5 per cent.

In the case of band spectra it was to be expected on theoretical grounds that distinct differences between isotopic molecules must reveal themselves, for some of the constituent lines arise from the rotations of the molecules and the vibrations of the component atoms, and these depend directly on the mass. By virtue of the extraordinary sensitivity of spectroscopic methods it has been possible not only to confirm by photographs of band spectra the existence of isotopes already found by the mass-spectrograph, but in recent years constituents have been observed in the band spectra of various elements which could only be interpreted by the assumption of hitherto unknown isotopes (e.g. ^{13}C, ^{15}N, ^{17}O, and ^{18}O) that remain invisible in mass-spectrograms owing to the small amounts of them present.

Moreover, the experiments on the artificial transmutation of the elements have introduced us to a large number of additional isotopes. But most of them are radioactive and do not occur in nature (see Chap. X); for this reason they are not included in Table 43. An exception has been made only in the cases of ^{3}T, ^{3}He, and ^{10}Be. Up to the present these elements are only represented among the atomic types prepared artificially, but as they are presumably stable there is some probability that they will be found in nature.

There is another reason, however, why the study of the artificial transformations of the atom is also very important for our understanding of the stable isotopes. When we realize artificially the transition of one atom into another and can measure the yield of energy that occurs in the process, we are in a position to calculate from this the relative masses of the two atoms to a higher degree of accuracy than is possible by measuring up mass spectra (see Chap. X, p. 124). From amongst the masses of the light atoms (up to oxygen) given in Table 43 only ^{1}H, ^{2}D, ^{4}He, ^{10}B, ^{12}C, and ^{14}N have been determined by mass-spectroscopic means, whereas the other masses have been calculated from the energy relations of the transformations of these elements into each other, produced artificially [4].

All the stable types of atoms that have been established by one of the methods mentioned are inserted in Table 43 with their mass-numbers. The precisional values of the atomic weights, or, what is the same thing, the deviations of the atomic weights from the integral values of the mass-numbers, have been introduced only for the elements up to argon, in order to keep the table within reasonable limits. For many of the heavy elements there are as yet available no precisional determinations of approximately the same degree of accuracy. As well as the mass-numbers, the table also contains the relative proportions of the isotopes present in the elements occurring in nature, and in many cases these can be derived with considerable accuracy from mass-spectrograms, by making use of photometric records.

When both the precisional values of the masses of its isotopes and also their quantitative relative abundance are known for an element, it is of course possible to calculate from this the combining weight of the element, and thus to test the values established by chemists in quite an independent way. If the greatest accuracy is required, it is necessary in this calculation to take account of the fact that the basis of the units of atomic weight in chemistry is not quite the same as that in physics. In chemistry the fictitious 'atomic weight' of the mixed element oxygen, which is in reality a mean value of the weights of the three oxygen isotopes corresponding to their relative abundance, is taken to be 16·0000, whereas in physics the value 16·0000 is ascribed to the mass of the oxygen isotope ^{16}O. Since the mixed element oxygen contains atoms of ^{18}O along with atoms ^{16}O in the relative proportions of 1 to 630 (^{17}O is so feebly represented that its influence can be neglected), the combining weight of the mixed element oxygen is, from the physical viewpoint, fixed at too low a value by the factor 0·9997. For this reason all the combining weights of mixed elements calculated on the basis of physical atomic weights must be multiplied by this factor in order that they may agree with the chemical scale of combining weights. In the last two columns of Table 43, for all elements for which sufficient data are already available for the purpose, the combining weights are given side by side as calculated on the one hand from the isotopic constitution, and on the other hand as determined directly by chemical means. It is seen that the agreement in most cases is excellent. It should be mentioned that in the case of several elements the calculation based on the isotopic constitution pointed to the fact that the combining weight determined by chemical methods was in need of revision.

The large amount of material on isotopes contained in Table 43 cannot yet be interpreted in its entirety from the theoretical point of view, but there is no doubt that the values of the relative abundance of the types of atom present in the individual elements give important clues concerning the stability of the relevant atomic nuclei (cf. Chap. X). Thus it is a very striking fact that elements with odd atomic number have on the average by no means so many isotopes as those elements with even atomic number. Only in the case of a single element of odd atomic number (potassium) has it been possible to detect a third isotope (^{40}K) with certainty (see p. 237), but its relative amount is so small (1 in 8,000) [5] that we can formulate the general rule that elements of odd atomic number never possess more than two isotopes present

TABLE 43

Isotopes of the Elements

Atomic number	*Element*	*Symbol*	*Mass-number*	*Atomic weight*	*Abundance in %*	*Combining weight (oxygen = 16)*	
						Calculated from isotopic composition	*Determined directly*
0	Neutron	*n*	1	1·0090	. .	. .	. .
1	Hydrogen	H	1	1·0081	99·98	1·0081	1·0078
	Deuterium	D	2	2·0147	0·02		
	Triterium	T	3	3·0171	. .		
2	Helium	He	3	3·0171	. .	4·003	4·002
	Helium	He	4	4·0039	100		
3	Lithium	Li	6	6·0167	7·9	6·9374	6·940
	Lithium	Li	7	7·0180	92·1		
4	Beryllium	Be	8	8·0078	(0·05)	9·0129	9·02
	Beryllium	Be	9	9·0149	99·95		
	Beryllium	Be	10	10·0164	. .		
5	Boron	B	10	10·0161	20	10·811	10·82
	Boron	B	11	11·0128	80		
6	Carbon	C	12	12·0036	99·3	12·008	12·01
	Carbon	C	13	13·0073	0·7		
7	Nitrogen	N	14	14·0073	99·62	14·008	14·008
	Nitrogen	N	15	15·0048	0·38		
8	Oxygen	O	16	16·0000	99·8	16·0000	16·0000
	Oxygen	O	17	17·0046	0·03		
	Oxygen	O	18	18·0038	0·16		
9	Fluorine	F	19	19·0049	100	19·000	19·000
10	Neon	Ne	20	19·9986	90·0	20·191	20·183
	Neon	Ne	21	. .	0·27		
	Neon	Ne	22	21·9985	9·73		
11	Sodium	Na	23	. .	100	. .	22·994
12	Magnesium	Mg	24	23·9938	77·4	24·33	24·32
	Magnesium	Mg	25	. .	11·5		
	Magnesium	Mg	26	. .	11·1		
13	Aluminium	Al	27	26·9911	100	26·985	26·97
14	Silicon	Si	28	27·9860	89·6	28·122	28·06
	Silicon	Si	29	28·9864	6·2		
	Silicon	Si	30	29·9845	4·2		
15	Phosphorus	P	31	30·9844	100	30·978	30·978
16	Sulphur	S	32	31·9812	96	32·05	32·06
	Sulphur	S	33	. .	1		
	Sulphur	S	34	33·9799	3		
17	Chlorine	Cl	35	34·9796	76	35·452	35·457
	Chlorine	Cl	37	36·9777	24		
18	Argon	A	36	35·976	0·31	39·953	39·944
	Argon	A	38	37·9753	0·06		
	Argon	A	40	39·9754	99·63		
			Mass-numbers (abundance in %)				
19	Potassium	K	39 (93·4); 40 (0·01); 41 (6·6)			39·096	39·096
20	Calcium	Ca	40 (96·76); 42 (0·77); 43 (0·17); 44 (2·30)			40·076	40·08

TABLE 43 (*cont.*)

Atomic number	*Element*	*Symbol*	*Mass-numbers (abundance in %)*	*Combining weight (oxygen = 16)*	
				Calculated from isotopic composition	*Determined directly*
21	Scandium	Sc	45 (100)	44·96	45·10
22	Titanium	Ti	46 (8·5); 47 (7·8); 48 (71·3); 49 (5·5); 50 (6·9)	47·91	47·90
23	Vanadium	V	51	..	50·95
24	Chromium	Cr	50 (4·9); 52 (81·6); 53 (10·4); 54 (3·1)	52·005	52·01
25	Manganese	Mn	55	..	54·93
26	Iron	Fe	54 (6·5); 56 (90·7); 57 (2·8)	55·84	55·84
27	Cobalt	Co	57 (0·17); 59 (99·83)	..	58·94
28	Nickel	Ni	58 (67·3); 60 (26·9); 61 (1); 62 (3·8); 64 (1)	58·71	58·69
29	Copper	Cu	63 (70); 65 (30)	..	63·57
30	Zinc	Zn	64 (50·4); 66 (27·2); 67 (4·2); 68 (17·8); 70 (0·4)	65·33	65·38
31	Gallium	Ga	69 (61·5); 71 (38·5)	69·71	69·72
32	Germanium	Ge	70 (21·2); 72 (27·3); 73 (7·9); 74 (37·1); 76 (6·5)	72·57	72·60
33	Arsenic	As	75 (100)	74·918	74·91
34	Selenium	Se	74 (0·9); 76 (9·5); 77 (8·3); 78 (24·0); 80 (48·0); 82 (9·3)	78·95	78·96
35	Bromine	Br	79 (50·0); 81 (50·0)	79·910	79·916
36	Krypton	Kr	78 (0·42); 80 (2·45); 82 (11·79); 83 (11·79); 84 (56·85); 86 (16·70)	83·76	83·7
37	Rubidium	Rb	85 (72·8); 87 (27·2)	85·45	85·48
38	Strontium	Sr	84 (0·5); 86 (9·6); 87 (7·5) 88 (82·4)	87·62	87·63
39	Yttrium	Y	89	88·91	88·92
40	Zirconium	Zr	90 (48); 91 (11·5); 92 (22); 94 (17); 96 (1·5)	91·24	91·22
41	Niobium	Nb	93 (100)	92·90	92·91
42	Molybdenum	Mo	92 (14·2); 94 (10·0); 95 (15·5); 96 (17·8); 97 (9·6); 98 (23·0); 100 (9·8)	95·95	95·95
44	Ruthenium	Ru	96 (5); 99 (12); 100 (14); 101 (22); 102 (30); 104 (17)	101·1	101·7
45	Rhodium	Rh	103 (100)	102·92	102·91
46	Palladium	Pd	102 (0·8); 104 (9·3); 105 (22·6); 106 (27·2); 108 (26·8); 110 (13·5)	106·54	106·7
47	Silver	Ag	107 (52·5); 109 (47·5)	107·87	107·880
48	Cadmium	Cd	106 (1·5); 108 (1·0); 110 (15·6); 111 (15·2); 112 (22·0); 113 (14·7); 114 (24·0); 116 (6·0)	112·2	112·41
49	Indium	In	113 (4·5); 115 (95·5)	114·80	114·76

TABLE 43 (*cont.*)

Atomic number	*Element*	*Symbol*	*Mass-numbers (abundance in %)*	*Combining weight (oxygen = 16)*	
				Calculated from isotopic composition	*Determined directly*
50	Tin . .	Sn	112 (1·1); 114 (0·8); 115 (0·4); 116 (15·5); 117 (9·1); 118 (22·5); 119 (9·8); 120 (28·5); 122 (5·5); 124 (6·8)	118·70	118·70
51	Antimony .	Sb	121 (56); 123 (44)	121·78	121·76
52	Tellurium .	Te	122 (2·9); 123 (1·6); 124 (4·5); 125 (6·0); 126 (19·0); 128 (32·8); 130 (33·1)	127·58	127·61
53	Iodine . .	I	127 (100)	126·904	126·92
54	Xenon . .	X	124 (0·08); 126 (0·08); 128 (2·30); 129 (27·13); 130 (4·18); 131 (20·67); 132 (26·45); 134 (10·31); 136 (8·79)	131·125	131·3
55	Caesium .	Cs	133 (100)	132·90	132·91
56	Barium . .	Ba	130 (0·16); 132 (0·02); 134 (1·72); 135 (5·7); 136 (8·5); 137 (10·8); 138 (73·1)	137·35	137·36
57	Lanthanum .	La	139 (100)	138·91	138·92
58	Cerium . .	Ce	140 (89); 142 (11)	140·13	140·13
59	Praseodymium	Pr	141 (100)	140·91	140·92
60	Neodymium .	Nd	142 (25·95); 143 (13·0); 144 (22·6); 145 (9·2); 146 (16·5); 148 (6·8); 150 (5·95) [6]	144·29	144·27
62	Samarium .	Sa	144 (3); 147 (17); 148 (14); 149 (15); 150 (5); 152 (26); 154 (20)	150·1	150·43
63	Europium .	Eu	151 (50·6); 153 (49·4)	151·90	152·0
64	Gadolinium .	Gd	155 (21); 156 (23); 157 (17); 158 (23); 160 (16)	156·9	156·9
65	Terbium .	Tb	159 (100)	158·19	159·2
66	Dysprosium .	Dy	161 (22); 162 (25); 163 (25); 164 (28)	162·5	162·46
67	Holmium .	Ho	165 (100)	164·91	163·5
68	Erbium . .	Er	166 (36); 167 (24); 168 (30); 170 (10)	167·15	167·2
69	Thulium .	Tu	169 (100)	168·91	169·4
70	Ytterbium .	Yb	171 (9); 172 (24); 173 (17); 174 (38); 176 (12)	173·2	173·04
71	Lutecium .	Lu	175 (98·5); 177 (1·5)	174·91	175·0
72	Hafnium .	Hf	176 (5); 177 (19); 178 (28); 179 (18); 180 (30)	178·4	178·6
73	Tantalum .	Ta	181 (100)	180·89	180·88
74	Tungsten .	W	182 (22·6); 183 (17·3); 184 (30·2); 186 (29·9)	183·94	184·0
75	Rhenium .	Re	185 (38·2); 187 (61·8)	186·2	186·31

TABLE 43 (*cont.*)

Atomic number	*Element*	*Symbol*	*Mass-numbers (abundance in %)*	*Combining weight (oxygen = 16)*	
				Calculated from isotopic composition	*Determined directly*
76	Osmium .	Os	186 (1·0); 187 (0·6); 188 (13·4); 189 (17·4); 190 (25·1); 192 (42·5)	190·30	191·5
77	Iridium . .	Ir	191 (33); 193 (67)	192·3	193·1
78	Platinum .	Pt	192 (0·8); 194 (30·2); 195 (35·3); 196 (26·6) 198 (7·2)	195·13	195·23
79	Gold . .	Au	197 (100)	..	197·2
80	Mercury .	Hg	196 (0·10); 197 (0·01); 198 (9·89); 199 (16·45); 200 (23·77); 201 (13·67); 202 (29·27); 203 (0·006); 204 (6·85)	200·60	200·61
81	Thallium .	Tl	203 (29·4); 205 (70·6)	204·41	204·39
82	Lead . .	Pb	204 (1·15); 206 (24·55); 207 (21·35); 208 (52·95) [7]	207·21	207·21
83	Bismuth .	Bi	209 (100)	..	209·00
90	Thorium .	Th	232 (100)	232·02	232·12
92	Uranium .	U	(234); 235 (0·4); 238 (99·6)	238·02	238·07

in comparable amounts. As an additional rule we can state that the number of isotopes even for elements of even atomic number never exceeds 10, and that the difference in mass between the lightest and the heaviest isotope amounts to no more than 12. For all elements apart from hydrogen and ^{3}He the mass even of the lightest isotope is at least double the value of the atomic number ($M \geqslant 2N$). The excess of the value of the mass above the double value of the atomic number is on the average greater, the greater the value of the atomic weight; thus for mercury the lightest isotope is already by 36 units heavier than $2N$.

If we also include the radioactive elements in our considerations, we can say that all integers from 1 to 238 are made use of by the mass-numbers of one or more types of atom, with the single exception of the number 5, for which no representative is yet known. Indeed, some of the numbers are associated with isotopes of several elements. Thus the main isotopes of argon, calcium, and a weak constituent of potassium all have the mass 40, and the three elements tin, xenon, and tellurium all possess atomic types of mass 124. Such atomic types of different elements which possess the same mass are called 'isobars'. In so far as precisional determinations of the masses of isobars are available, it has been found that they are in very exact agreement (cf. ^{3}T and ^{3}He;

^{10}Be and ^{10}B). Moreover, for any future theory of the structure of the atomic nucleus, the following result found for light atoms will be of importance, to wit, those types of atoms whose mass-numbers amount to a multiple of 4 show no excess in atomic weight above integral values (relative to $^{16}O = 16{\cdot}000$), whereas the intervening atomic nuclei do show such an excess in mass, and hence they appear to have a less stable structure.

Finally, it is a particularly surprising fact that the combining weights of all the elements appear to be natural constants, in spite of the large number of isotopes of which many of them are composed. (On this point see Chap. XXI, p. 200 et seq.)

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XX

THE SEPARATION OF ISOTOPES

In Chapter XV we have discussed the attempts of chemists to separate from each other certain pairs of radio-elements such as radium and mesothorium, thorium and ionium, or radium D and lead. These constitute the first endeavours made for the purpose of separating isotopes. All of them yielded a negative result, and in consequence they led to the formulation of the idea of isotopy. A much wider field of inquiry was opened up by the establishment of the phenomenon of isotopy for the ordinary elements (see Chap. XIX), because in this case there is no limitation to the amount of material available for experiment, and also no limitation in time to be considered owing to spontaneous disintegration during the application of the experimental method used. In spite of the extraordinarily varied experimental conditions to which they have been subjected during many physical and chemical investigations, not even a partial separation of any of the 'mixed elements' (see p. 199) has been noted. From this it follows that, in general, a separation of ordinary isotopes cannot be an easy matter. Nevertheless, before the discovery of the phenomenon of isotopy, no systematic experiments in this direction had been undertaken. Apart from its theoretical interest, accurate knowledge of the conditions under which isotopes can be separated is also necessary, because of the fact that important constants such as the combining weights or the density of mercury can be influenced in a very disturbing manner by such a separation.

Actually there is an element whose isotopes can be separated so readily that not only are the laboratory methods of separating them relatively simple, but the mixing-ratio of the two isotopes in nature also shows fluctuations. The element in question is hydrogen. The properties of 'heavy hydrogen' or 'deuterium' (atomic weight = 2·0147) differ so strongly from those of the much more abundant ordinary hydrogen (atomic weight = 1·0081) that the two components can be easily separated from each other [1] by fractional distillation of liquid hydrogen, by methods of diffusion and also by the particularly simple and convenient method of the electrolysis of water. In what follows, when we are speaking of the methods of separating isotopes, we shall take no further account of this quite exceptional case, in which the one isotope is twice as heavy as the other.

It should be stated in advance that we can only assume the success of a real separation of isotopes if they were available at the beginning of the experiment as a completely uniform mixture. This condition is always satisfied (see Chap. XXI) for the isotopic types of atoms of the elements which occur in nature, but not for the isotopic radioactive substances. Thus, radiothorium extracted from minerals is always accompanied by thorium, with which it is isotopic; but it can readily be prepared free from thorium by separating its immediate parent element mesothorium from the thorium, and awaiting the growth of fresh radiothorium from the mesothorium. In a similar manner all isotopic radio-elements can be obtained in the separated condition if their parent elements are separable, and their reformation does not demand too long a period of time. But it is clear that in these cases, which are based on the divers parentage of the isotopes, we must not speak of a separation of the isotopes, since the separation only has reference to their parent elements, which are not isotopes. The problem assumes a somewhat different complexion when, as a result of the diversity of life of two mixed isotopes, it is possible to obtain one of them in the pure state. This case arises, for instance, with the active deposit collected from the atmosphere. At first it contains the two isotopes radium B and thorium B, but after one day practically only thorium B is left, since the radium B has meanwhile decayed almost completely into RaD. For this reason it is hardly correct to speak here of a separation of the isotopes, because one of the isotopes entirely vanishes.

Nevertheless, there are also certain less clear cases in which a separation of mixed radioactive isotopes appears to have been really successfully carried out. Thus it has been observed that the ratio thorium B:thorium C as compared with that of radium B: radium C increases when these four elements are adsorbed simultaneously by iron hydroxide. Either the two isotopic B-products or the likewise isotopic C-products must therefore have been adsorbed in different proportions. If, in spite of this, we cannot here speak of a separation of the isotopes in the true sense, the reason is to be found in the fact that the condition of completely uniform mixing before the commencement of the experiment is not satisfied. The B-products and still more the C-products have a great tendency to go into solution with the properties of colloidal particles, and the amount of the colloidal and the molecularly disperse portions depends on the time during which the solvent medium has been allowed to act. Owing to the different half-value periods, the duration of this action of the solvent is not the same for the thorium and the radium products, and since the adsorption is stronger for colloidal particles than for molecularly disperse ones, the result is a displacement of the concentration ratio.

Correspondingly, one process for the concentration of isotopes produced artificially is based on the fact that these freshly produced atomic types are in another state of chemical binding than the original material [2]. Thus when an organic substance that contains iodine in combination with carbon is bombarded by neutrons, radioactive isotopes of iodine are produced by the capture of neutrons. In this process, however, the molecular binding is severed and the radioactive iodine atoms are not united with carbon. Because of this, it is possible to obtain the active iodine in a very concentrated form by precipitation of the free iodine with silver, or by shaking up the organic liquid well with water, when all the active iodine ions are concentrated in the water [3]. Also, by electrolysis in the liquid or gaseous phase, we can in this way readily concentrate the active isotopes of iodine [4]. These processes do not constitute a separation of the isotopes in the true sense, since the condition of initially complete uniformity in distribution is not satisfied, and it is just this inhomogeneity that is utilized in practice for the concentration of the isotopes. On the other hand, the method of using radio-elements as indicators (see Chap. XVIII) is based essentially on their inseparability, and it is therefore clear that we must here be very careful to ensure that the process of mixing with the indicator is carried out in the molecularly disperse solution, or in the liquid or the gaseous state.

The first method to achieve a partial separation of isotopes in weighable amounts was that of 'ideal distillation'. It is based on the following consideration:

When a liquid is in equilibrium with its saturated vapour, a definite number of molecules of the vapour strike the surface of the liquid in unit time, and are captured; this number must be equal to the number of molecules that pass from the liquid to the vapour phase in the same time. The velocity of this process of kinetic exchange depends on the mean velocity of the molecules of vapour, and likewise on the velocity of the molecules in the liquid. The lighter (mass m_1) of two isotopes will therefore be in a state of more vigorous exchange than the heavier isotope (mass m_2). Now according to the laws of the kinetic theory the product mv^2 must be constant, so that the velocities will be related inversely as the square roots of the masses. From this it follows that in the unit of time $\sqrt{\frac{m_2}{m_1}}$ times as many molecules of the lighter isotope as of the heavier isotope will pass over from the surface into the vapour phase and, conversely, will be condensed from the vapour phase. In the ordinary process of distillation the number of molecules condensed in the distillate is only a small fraction of the number of molecules that take part in the kinetic interchange between the liquid and the vapour in the same time. Thus, although in unit time more molecules of the lighter isotope leave the liquid, this isotope does not become more concentrated in the distillate, since conversely and at the same time more molecules of the lighter isotope condense out from the vapour phase. The average composition of the vapour is the same as that of the liquid. But if we conduct the process along irreversible channels by preventing the molecules which leave the liquid from being returned to it, it is then possible to concentrate the lighter isotope in the distillate. If all the molecules that have left the liquid are held fast before their direction is reversed by collisions with other molecules, then the proportion in which the isotopic molecular types exist in the distillate must be equal to their ratio in the liquid, multiplied by the ratio of their average velocities. It follows that $\frac{c_1}{c_2} = \sqrt{\frac{m_2}{m_1}}$ times as much of the lighter isotope will be condensed as of the heavier isotope. We see, then, that although the *vapour pressures* of two isotopes, except at the lowest temperatures, are indistinguishably identical, it is possible to effect a partial separation by utilizing the fact that their *velocities of vaporization* are different. The degree of separation can be increased by repetition of this process of 'ideal distillation', just as in the case of other fractional distillations.

From what has been said it is clear that various conditions must be fulfilled in order that the method may be a practicable one. In the first place the vapour pressure must be kept so low that the mutual collisions of the molecules can be neglected, as they effect a recoil of the escaping molecules back into the liquid. Furthermore, the surface upon which the vaporized molecules strike must be of such a nature that all the molecules are held by it, and none of them are reflected. This is attained most conveniently by the introduction of a strongly cooled glass surface opposite to the liquid surface. Finally, care must be taken to ensure that the composition of the surface of the liquid does not change relatively to the interior of the liquid; for if the kinetic exchange between the surface and the interior of the liquid does not take place sufficiently rapidly, the escape of the lighter isotope will result in a concentration of the heavier isotope in the surface layers, and this will prevent any further concentration of the lighter isotope in the space above the liquid. This last condition restricts the applicability of this method of separation to liquids, and renders it useless for solid bodies.

The first success achieved by this process of ideal distillation was in the partial separation of the isotopes of mercury. The apparatus used for this purpose is depicted in Fig. 46. 300 c.c. of mercury were introduced into the intervening space between the inner and outer walls of a Dewar vessel, the space being then highly evacuated and the interior of the flask *A* filled with liquid air. The mercury was heated to about 45° C. by means of the oil-bath *C*, so that its vapour pressure was about 0·01 mm. and the amount which vaporized per hour per square centimetre of its surface was about 0·35 c.c. This vaporized mercury froze on the glass wall of the flask *A*. After about one-quarter of the amount of mercury had been condensed in this way, the experiment was interrupted, and the residual heavier mercury was transferred to the evacuated vessel *E* by opening the stopcock *D*, whence it was allowed to flow from the apparatus by way of the stopcock *G*. The next step was the removal of the liquid air from the vessel *A*, so that the mercury that had condensed on it could thaw, after which it was removed from the apparatus in the same way as that used for the removal of the residue from the distillation. By carrying out this process repeatedly it was possible to divide an initial quantity of 2,700 c.c. of mercury into a number of fractions, some of which had a greater and some a lesser specific gravity than normal mercury. The heaviest fraction (volume = 0·2 c.c.) had a density of 1·00023 (relative to Hg = 1), and the lightest fraction (likewise 0·2 c.c.) a density of 0·99974, so that the difference in density between the heaviest and the lightest samples of mercury amounted to about 0·05 per cent. For equal volumes the conductivities of the two samples were indistinguishably identical, whereas the combining weights differed by almost 0·1 unit, as was to be expected. The combining weight of the heaviest fraction was 200·632, and that of the lightest fraction 200·564, whereas the combining weight of ordinary mercury is 200·61.

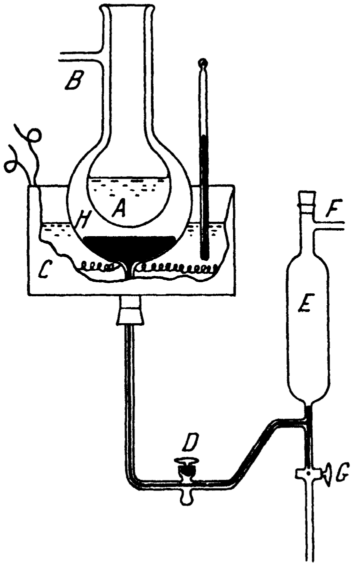

Fig. 46. Apparatus for the Partial Separation of the Isotopes of Mercury.

It has also been possible to effect a partial separation of the isotopes of chlorine by the same process, and using a 7N aqueous solution of HCl for the ideal distillation. The difference in the combining weights of the two chlorine fractions amounted to a little more than two units in the second decimal place, corresponding to a displacement of the mixing ratio of the two isotopes $^{35}\mathrm{Cl}$ and $^{37}\mathrm{Cl}$ of approximately 6 per cent.

A partial separation of the isotopes of zinc has also been carried out in this manner. Experiments performed in an attempt to displace the mixing ratio of the isotopes of potassium were of particular interest, because it was possible from them to draw conclusions as to the isotope responsible for the radioactivity of potassium. By use of the method of ideal distillation for the mixed element potassium, which consists of 93 per cent. of ^{39}K, 7 per cent. of ^{41}K, and 0·01 per cent. of ^{40}K, a heavier fraction was obtained. The radioactivity of this was found to be greater by 4·4 per cent. than that of ordinary potassium, which showed that the radioactivity of potassium is due to a heavier isotopic constituent. Atomic weight determinations of the heavier fraction revealed an increase in the concentration of ^{41}K by as much as 7·3 per cent. [5]. From this it follows that the radioactive isotope is less readily separated than ^{41}K, and that accordingly it must have the mass-number 40.

Experiments on fractional diffusion have been more effective than those of distillation for the separation of isotopes. When a mixture of two isotopes in the vapour or gaseous state is allowed to flow through a narrow opening, the lighter isotope will strike the opening more frequently than the heavier isotope, in consequence of its greater molecular velocity. As a result of this, the probability of the passage of the lighter isotope through the opening will be $\sqrt{\frac{m_2}{m_1}}$ times greater than that of the heavier isotope, where m_1 and m_2 are their respective masses. Now if care be taken to ensure that the molecules are caught on the other side of the opening, and so prevented from streaming back into the vessel, the lighter isotope will be concentrated in the condensed fraction for the ideal case in the ratio $\sqrt{\frac{m_2}{m_1}}$ by this method also.

A certain amount of success was achieved by a method of diffusion with the isotopes of neon as long ago as 1913, at the time of the discovery of the phenomenon of isotopy. Here the concentration was effected by the use of porous clay pipes. More definite results were obtained later by the application of a similar process to mercury, in which the diffusion took place through punctured platinum foils. The method was also applied with some success in the case of chlorine, by allowing HCl to diffuse through clay pipes. But it was not until 1932 that the principle became sufficiently far developed to make it possible to effect almost complete separation of gaseous isotopes in a single operation. The apparatus consists of 24 components of separation, each member consisting of two clay tubes and a mercury condensation pump (cf. Fig. 47, in which only four of the constituent parts are drawn, the dividing line between each being indicated by a dotted line). One-third of the gas that streams through the first clay tube of a separating component is drawn off by the pump of the right-hand neighbouring member, and in this process the lighter isotope is somewhat concentrated in this fraction. The gas remaining behind streams through the second clay tube, and a second third of the gas is drawn through its walls by the pump of the separating component under consideration, so that only the final third passes on to the left-hand component, in which the same process is repeated. The connexions are so arranged that the lighter isotope is progressively concentrated in a continuous stream from left to right through the whole apparatus, whereas the heavier isotope is progressively concentrated in a corresponding manner from right to left. Details of the connexions and of the disposition of apparatus at the two ends of the series of components can be seen in Fig. 47. The experiments have shown that in the case of neon the apparatus need only operate for 8 hours in order to obtain a few cubic centimetres of a gaseous mixture in which the isotopes of neon are present in almost equal proportions, whereas the intensity ratio ^{20}Ne:^{22}Ne has the value 9:1 in neon as it occurs in nature. A recently perfected model of the apparatus, consisting of 48 separating components and fitted with larger pumps, permits of still quicker and more efficient working of the system, and has been found especially valuable in the preparation of heavy hydrogen in the pure state [6]. For the purpose of separating the isotopic mixture it is also possible, by means of a special construction of the pumps, to utilize the diffusion of the mixture in the mercury vapour of the pumps, and to dispense with the pipe-clay tubes entirely.

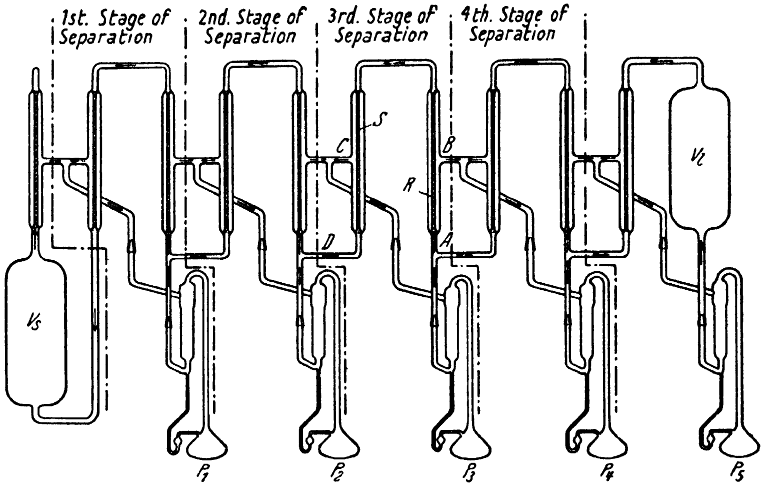

FIG. 47. Apparatus for the Separation of Isotopes by Diffusion.

There are several other methods of separation available which deserve brief mention because of their theoretical interest. Thus the positive-ray method, by means of which isotopes can be recognized by their separation, can also be used under certain circumstances for the preparation of the isotopes in the pure state. When the problem presented itself of examining separately the effect of the bombardment of the individual isotopes of lithium with light and with heavy hydrogen nuclei, films of metallic lithium consisting respectively of only ^{6}Li or ^{7}Li were prepared by resolving the rays from lithium in accordance with the principle used in the mass-spectrograph.

Since isotopes are practically identical in their chemical properties, the most efficient methods used for separating them are based on differences in their masses, and for this reason the processes so far described differ fundamentally from the methods applied for the separation of substances in other branches of chemistry. Nevertheless, in certain circumstances it is possible to effect a separation of isotopes also by normal distillation. On theoretical grounds it is to be expected that isotopes evince sensible differences in their vapour pressures at very low temperatures. If account be taken of zero-point energy, it is found that at low temperatures the lighter isotope must have the higher vapour pressure, whereas if there is no zero-point energy it will have the lesser vapour pressure. When neon is fractionally distilled at a temperature of —248·4° C., neon of unmistakably smaller density than the normal value is obtained in the more volatile portion, whereas a heavier fraction remains in the residue, so that the lighter isotope must have the greater vapour pressure. The significance of this result is primarily that it proves by means of an *experimentum crucis* the necessity for the acceptance of zero-point energy; but for the practical separation of isotopes, of course, this elaborate process can be dispensed with.

On theoretical grounds it is further to be expected that the separability of the hydrogen isotopes does not constitute a completely isolated example of separation by simple chemical and electrochemical processes. For the other light elements there also exists, albeit to a very much smaller degree, the possibility of a separation of their isotopes on the basis of their chemical or electrochemical dissimilarity. Such experiments have been carried out quite recently with lithium, carbon, nitrogen, and oxygen. In the case of carbon (isotopes ^{12}C and ^{13}C) use was made of the different reaction velocities of the two types of CO_2 with water; by using an enzyme as catalyser and a complicated counter-current scrubbing apparatus, the content of ^{13}C in one fraction was increased from 1·06 per cent. to 1·36 per cent. [7]. For the separation of the nitrogen isotopes ^{14}N and ^{15}N use was made of the reaction

$$^{15}NH_3 + {}^{14}NH_4^+ \rightleftarrows {}^{14}NH_3 + {}^{15}NH_4^+.$$

The ratio of ^{14}N to ^{15}N was found to have been reduced from 250 to 234 [8]. The vapour pressures of the ^{16}O and ^{18}O varieties of water are sufficiently different to ensure that, when a fractional distillation is carried out in the above-mentioned fractionation column, a threefold increase in concentration of the heavier isotope can be effected after the lapse of eight days [9]. Finally, by virtue of differences in the electrode potentials of ^{7}Li and ^{6}Li it has been possible to effect a change in their isotopic ratio from 11·6 to 5·1 [10].

In a few specially favourable cases it has been possible to effect a separation of isotopes by photochemical methods. Since carbon and oxygen are practically pure elements, and chlorine consists of the two atomic types ^{35}Cl and ^{37}Cl, it follows that there will be three different kinds of phosgene molecules: $CO^{35}Cl^{35}Cl$, $CO^{35}Cl^{37}Cl$, and $CO^{37}Cl^{37}Cl$. In the absorption spectrum different lines will correspond to these three types of molecules. For a certain triplet the component line of shortest wave-length has the value 2,816·179 A. and is due to the molecule $CO^{35}Cl^{35}Cl$. It is possible to produce light of exactly the same wave-length by removing all other wave-lengths from the spectrum of a spark between aluminium electrodes by means of light filters. As is well known, only molecules that absorb light are broken down photochemically, so that a decomposing action of this filtered light is to be expected only for this one kind of molecule. By raying the phosgene mixture in this way, it was in fact found that chlorine was liberated, and its combining weight was 35·429 instead of the normal value 35·457. It was thus shown that the filtered light had decomposed mainly those molecules that contained two chlorine atoms of mass 35. It was not to be expected that the action would be restricted exclusively to this type of molecule, since, in addition to the lines, the absorption spectrum shows also a continuous background. A similar partial separation of isotopes has also been carried out by the illumination of mercury vapour with filtered light [11]; mainly the excited isotopes were oxidized, and so removed from the gaseous phase.

On the basis of theoretical considerations various other methods have also been tried out, but they have not yet led to successful results. Thus in many cases the method of diffusion in aqueous solutions has been applied, as in attempts to separate the two isotopes of uranium. But for diffusion in liquids the advantage of greater molecular velocity possessed by the lighter isotope is compensated by the numerous collisions with the molecules of the liquid, by virtue of which the lighter isotope more readily loses its direction. Moreover, in aqueous solution the difference in the masses and hence also in the mobilities of the isotopic particles can be substantially diminished by the effect of hydration. An appreciable relative speeding up of the particles of the lighter isotope has in fact never been observed. The same thing holds for the mobility of the ions in solutions.

A spontaneous separation of the isotopes might be expected in the case of the mixed elements occurring in nature as a consequence of the effect of the gravitational field of the earth. The lighter gases in the atmosphere might be expected to become concentrated relatively to the heavier ones at higher levels, and it might likewise be assumed that at great heights the ratio of the two neon isotopes would be displaced in favour of the lighter isotope; thus at a height of 10 km., instead of 91 per cent. ^{20}Ne and 9 per cent. ^{22}Ne, there should be present 92 per cent. ^{20}Ne and only 8 per cent. ^{22}Ne. But such a displacement of the molecular composition of the atmosphere is only to be expected if we suppose that the distribution of the molecules in the gravitational field can take place strictly in accordance with the laws of diffusion, and without disturbance by convection currents. These last, however, really exert a preponderating influence within the atmosphere. Similar considerations hold validity for the dependence of the isotopic ratio on depth in the case of mixed elements from the ocean depths. NaCl from the deepest region of the ocean, about 10 km., should be by so much richer in ^{37}Cl than in ^{35}Cl that the combining weight of the chlorine contained in it should have the value 35·6 instead of 35·46. But here, also, the disturbance of the gravitational distribution by ocean currents is so strong that it has not been possible to detect the effect even qualitatively.

In principle, the same effects as are produced by gravitational forces should also be attainable by centrifugal forces. For the earth a rotational velocity of 1 km./sec. corresponds to a difference in height of about 40 km. It has not yet been possible to construct centrifuges of such efficiency, and at the same time so completely to exclude disturbances likely to arise from convection, as to render practicable a separation of isotopes by this means.

In view of the great difficulties with which the separation of isotopes taxes the equipment even of a modern laboratory, it is hardly to be expected that any natural chemical processes whatsoever, that take place on this planet, will be able to bring about an appreciable displacement of the isotopic ratio of an element, always excepting, of course, the case of hydrogen. In complete accord with this is the fact mentioned elsewhere (pp. 186, and 200–202), that the combining weights of the chemical elements are natural constants. Biological processes also appear to be ineffective in bringing about a separation of isotopes. Thus the isotopic composition of the element copper, extracted from the blood of the horseshoe crab (*limbus polyphemus*), has been the subject of careful examination, but the copper so obtained was found to have the same combining weight as ordinary copper [12].

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XXI

MIXED ELEMENTS AND PURE ELEMENTS. COMBINING WEIGHTS AND ATOMIC WEIGHTS

As we have seen in Chapter XIX, Dalton's hypothesis, that a chemical element consists only of one quite definite type of atom, has validity only for very few elements. Even with these, e.g. helium, it is very well possible that more sensitive methods will reveal the existence of isotopes (cf. Tables 43 and 56). In the case of the majority of the elements it is already known that they are composed of several isotopic types of atoms. As a result of the chemical inseparability of various radioactive substances (see p. 148) it became clear for the first time that the impossibility of resolving a substance chemically may not constitute proof that its atoms are all of one type, and this conjecture has been completely confirmed by investigations with positive rays. In this way our theoretical ideas on the nature of a chemical element have been strongly modified, since for over a hundred years one had regarded as certain the foundations of atomistics due to Dalton, according to which the number of kinds of atoms was equal to the number of the chemical elements. But the practical significance of this new knowledge is very small for the subject of chemistry, as is shown by the fact that the complex nature possessed by so many elements was so long overlooked by chemists, and was first disclosed by researches so far removed from chemistry as the investigations on positive rays. Hence it is that the regularities in chemistry embodied in the periodic classification must continue to be built up on the conception of the chemical element.

We have already had occasion to discuss the fluctuations in the combining weight of lead obtained from different sources (see p. 150). From the chemical viewpoint, however, these kinds of lead, in which the isotopic atom types are mixed in various proportions, are unitary elements. Their purification and the determination of their 'atomic weight' could be carried out by the usual methods. Such an element consisting of several isotopes is termed a 'mixed element'—an expression we have already used—and it is preferable to speak of its 'combining weight' rather than of its atomic weight, since in the case of chlorine, for example, the relative weights of the atoms contained in it are 35 and 37, whereas its combining weight is 35·457. An element that consists of only one type of atoms is called a 'pure element', and it is associated with a constant atomic weight which indicates directly the weight of its atoms relative to oxygen = 16·0000. When we speak generally of a 'chemical element', it is not clear whether we are dealing with a pure element, or with a mixed element such as lead or chlorine.

In view of present-day knowledge we can no longer simply say that the chemical elements cannot be further resolved; it is possible to separate the various types of atoms present in them, and thus to resolve them into substances of different kinds. To-day we may define an element conveniently in the following way: *A chemical element is a substance which is not resolved into simpler substances by the usual chemical processes.* The fact that the elements are not resolved chemically retains its validity for all practical purposes, and here we have the reason why these substances which persist in all chemical reactions rightly bear the name of chemical 'elements'. For other than chemical experiments the same substances do not represent something that cannot be resolved. By means of certain artifices we can bring about a separation of the isotopic atom types (see Chap. XX), and by others a disruption of the atoms (see Chap. X); but since neither of these processes falls within the ambit of normal chemical operations, this compositeness of the elements, which is present in a dual sense, can be neglected in the whole of the vast field of practical chemistry. For practical and didactic purposes the above definition is in general sufficiently precise, since almost all the successful methods of separating isotopes are typically mechanical ones, inasmuch as in them we make use of differences in mass and not of a difference in their chemical properties. Nevertheless, it has been possible to attain by photochemical means and by exchange reactions a partial separation of the isotopes of a few light elements. Moreover, for the isotopic atom types of hydrogen the difference of their chemical properties also is so great that we should certainly have been tempted to consider them as two different chemical elements, had it not been for the fact that the almost complete identity of the chemical properties of the isotopes of all the rest of the elements has led us to regard the nuclear charge as the primary characteristic of an element.

This more exact and more fundamental definition, even though it is more remote from the original chemical meaning of the element concept, can be formulated in the following way:

A chemical element is a substance of which all the atoms have the same nuclear charge. Examples: helium (nuclear charge 2), chlorine (nuclear charge 17), lead from an arbitrary mineral (nuclear charge 82), lead from disintegrated radon (nuclear charge 82).

A pure element is an element that consists of only one type of atom. Examples: helium (atoms of mass 4·002), lead from radon (atoms of RaD, which emits β-rays, of mass 210).

A mixed element is an element that consists of several types of atoms. Examples: chlorine (atoms of mass 35 and 37), lead from galena (atoms of mass 204, 206, 207, and 208).

In the thermodynamic sense a pure element is thus a unitary substance, whereas a mixed element is not. But since we consider both mixed elements and pure elements as chemical elements, we see that the necessity of retaining the systematics of chemistry in spite of the discovery of isotopy has compelled us to free the concept of the chemical element from the thermodynamic concept of matter. The members of the periodic classification of the elements are chemically unresolvable, but in general they are not thermodynamically unitary substances.

In the domain of the radio-elements, for which the isotopic atom types occur separately in nature, in contrast to the ordinary elements, it is usual to enter the most long-lived type in the periodic classification, for with it the chemical properties can be observed most readily; e.g. thorium is taken as element 90, and not, say, uranium X_1 (cf. Tables 40 and 41 with Table 38). Only occasionally, and for special purposes, do we enter all the types of the radio-elements also in the tables of the periodic classification.

The question as to whether a chemical element is a pure or a mixed element is of no practical importance to the chemist, and he is also interested more in the combining weight than in the weight of the individual atom types of an element. Thus, although for scientific purposes a complete table of the 'atom types' is important, the old-time table of the 'chemical elements' and of their 'combining weights' has not lost its significance. Since we have tabulated the atom types of the radio-elements already in Table 38, p. 152, and the atom types of the inactive elements so far as they are at present known in Table 43, p. 182, there only remains to be given here one embodying the chemical elements and their combining weights (Table 44). The arrangement is the one usual in chemistry, the elements being entered in the alphabetical order of their symbols. We have also given the atomic number of each of the elements after its combining weight; with the help of this number we can immediately find the position of the particular element in the periodic classification (see Tables 40 and 41), as well as its atomic composition (see Table 43).

Since the combining weights of the mixed elements are only average values obtained from the weights and the proportions of the isotopic atomic types contained in them, the question arises as to whether they can still be regarded as natural constants. It appears that we can without reservation answer this question in the affirmative, so long as we confine ourselves to the ordinary inactive elements. For these it has not been possible hitherto to establish with certainty fluctuations in their combining weights, even when material from quite different sources has been investigated, with one solitary exception. This is hydrogen, for which a displacement of the mixing ratio of the isotopes

TABLE 44

Combining Weights and Atomic Numbers of the Chemical Elements

Symbol	*Name of element*	*Combining weight*	*Atomic number*	*Symbol*	*Name of element*	*Combining weight*	*Atomic number*
Ag	Silver	107·880	47	N	Nitrogen	14·008	7
Al	Aluminium	26·97	13	Na	Sodium	22·994	11
Ar	Argon	39·944	18	Nb	Niobium	92·91	41
As	Arsenic	74·91	33	Nd	Neodymium	144·27	60
Au	Gold	197·2	79	Ne	Neon	20·183	10
B	Boron	10·82	5	Ni	Nickel	58·69	28
Ba	Barium	137·36	56	O	Oxygen	16·0000	8
Be	Beryllium	9·02	4	Os	Osmium	191·5	76
Bi	Bismuth	209·00	83	P	Phosphorus	30·978	15
Br	Bromine	79·916	35	Pb	Lead	207·21	82
C	Carbon	12·01	6	Pd	Palladium	106·7	46
Ca	Calcium	40·08	20	Pr	Praseodymium	140·92	59
Cd	Cadmium	112·41	48	Pt	Platinum	195·23	78
Ce	Cerium	140·13	58	Ra	Radium	226·05	88
Cl	Chlorine	35·457	17	Rb	Rubidium	85·48	37
Co	Cobalt	58·94	27	Re	Rhenium	186·31	75
Cr	Chromium	52·01	24	Rh	Rhodium	102·91	45
Cs	Caesium	132·91	55	Rn	Radon	222	86
Cu	Copper	63·57	29	Ru	Ruthenium	101·7	44
Dy	Dysprosium	162·46	66	S	Sulphur	32·06	16
Er	Erbium	167·2	68	Sb	Antimony	121·76	51
Eu	Europium	152·0	63	Sc	Scandium	45·10	21
F	Fluorine	19·00	9	Se	Selenium	78·96	34
Fe	Iron	55·84	26	Si	Silicon	28·06	14
Ga	Gallium	69·72	31	Sm	Samarium	150·43	62
Gd	Gadolinium	156·9	64	Sn	Tin	118·70	50
Ge	Germanium	72·60	32	Sr	Strontium	87·63	38
H	Hydrogen	1·0078	1	Ta	Tantalum	180·88	73
He	Helium	4·002	2	Tb	Terbium	159·2	65
Hf	Hafnium	178·6	72	Te	Tellurium	127·61	52
Hg	Mercury	200·61	80	Th	Thorium	232·12	90
Ho	Holmium	163·5	67	Ti	Titanium	47·90	22
In	Indium	114·76	49	Tl	Thallium	204·39	81
I	Iodine	126·92	53	Tu	Thulium	169·4	69
Ir	Iridium	193·1	77	U	Uranium	238·07	92
K	Potassium	39·096	19	V	Vanadium	50·95	23
Kr	Krypton	83·7	36	W	Tungsten	184·0	74
La	Lanthanum	138·92	57	X	Xenon	131·3	54
Li	Lithium	6·940	3	Y	Yttrium	88·92	39
Lu	Lutecium	175·0	71	Yb	Ytterbium	173·04	70
Mg	Magnesium	24·32	12	Zn	Zinc	65·38	30
Mn	Manganese	54·93	25	Zr	Zirconium	91·22	40
Mo	Molybdenum	95·95	42				

takes place much more readily than for the other elements. For example, water from the Dead Sea or from the leaves of a tree contains a little more of the heavy variety of water than does rain-water. But copper and chlorine from widely separated sources of origin, iron, nickel, silicon, and chlorine from terrestrial minerals and from meteorites, mercury from minerals of very different geological age and geographical occurrence, all of these showed complete constancy in their combining weights within the limits of experimental error (cf. Chap. XIX, where this question has already been mentioned). The ratio of the isotopes ^{16}O and ^{18}O in stone meteorites is the same as in terrestrial oxygen; this was shown by determinations of the specific gravity of water made from these sources of oxygen, and by means of mass-spectrograms [1]. From these facts we must conclude that the atomic types of the elements were already formed and had had opportunity for complete mixing before the solidification of the planets. This conclusion is in best agreement with the assumption, which for other reasons seems probable, that our solar system was initially in a fluid magma or gaseous state.

But in the framework of these ideas we must expect that the constancy in the combining weight will be absent when a new formation of atom types has taken place since the solidification of the earth's crust. This case has arisen exclusively with the radio-elements. Thus lead which has been formed in uranium minerals during the course of geological time has had little or no opportunity of mixing with ordinary lead, and in accordance with this we find here those fluctuations in the combining weight which we have discussed in Chapter XV. An analogous case is the low combining weight of strontium formed by the disintegration of rubidium (see p. 237). The deviations in the cases of lead, thorium, and strontium of radioactive origin are, together with those achieved for a number of elements by an artificial separation of the isotopes (see Chap. XX, p. 187), the only fluctuations that it has been possible so far to establish. In all other cases—always with the exception of hydrogen—with the present-day attainable accuracy, the combining weights of the chemical elements are natural constants, which have not lost any of their practical importance as a result of the recognition of the complex nature of the mixed elements.

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XXII

THE PREPARATION OF RADIOACTIVE SUBSTANCES

It will be clear from what has already been said about the chemical nature of radioactive substances that they do not constitute a group of elements with the same, or even with similar chemical properties. Their physical behaviour, the disintegration of their atoms, is the only thing they have in common. From the chemical point of view they differ greatly from one another, and this applies even to elements such as radium and radon which are very closely related genetically. For this reason, then, no single process can be applied to the production of one and all of them, and it is not possible to state certain group reactions as, say, in the case of the rare earths or the alkali metals.

Moreover, owing to their radioactivity and consequent instability, we are here faced with special difficulties such as we meet in no other branch of chemistry. An inactive substance that has once been freed from foreign chemical elements remains permanently pure, provided it is suitably stored. It would strike a chemist as being very remarkable if chemical elements that he had carefully removed were to reappear after the lapse of a few hours or weeks. But with radioactive substances this is always occurring, with some more quickly than with others. Of course, when the substance is regenerated after an interval of a few hours or a few minutes, the amounts formed in this time are never weighable nor visible, but are only rendered evident by the radiation they emit. In the ordinary chemical sense the preparations have remained pure, and it is impossible to recognize the admixture of a foreign element either by means of reagents or with the aid of a spectroscope. Nevertheless, the electroscope reveals to us that the substance which had been removed is again present in the material.

In the preparation of radioactive substances in a state of purity it is therefore necessary to differentiate between the terms 'chemically pure' and 'radioactively pure'. The methods we apply will differ greatly according to the purpose we have in hand. 'Chemically pure' thorium, to take one example, may contain several radioactive disintegration products as impurities without its utility being adversely affected. Conversely, a preparation of radium which is pure from the radioactive viewpoint (e.g. free from mesothorium) may, for many radiological investigations, contain quite appreciable amounts of barium. In the *chemical* sense we can obtain only very few of the radioactive substances in the pure state; besides uranium and thorium, only protoactinium, radium, radon, and radium D can be obtained in visible quantities. Nevertheless, we are rarely concerned with the chemical purity of these substances, for their most valuable feature is their radioactive properties, which lose little or none of their effectiveness by the admixture of inactive material. Thus chemically pure radium chloride (i.e. 100 per cent. $RaCl_2$) has so far only been prepared on three occasions (in Paris, in Vienna, and in Munich), and for scientific purposes. We also rarely require complete purity in the radioactive sense, for an 'impurity', especially of the disintegration products of the substance itself, is quite welcome in practice, because in this way the activity of the substance (e.g. mesothorium or radiothorium) increases. In scientific investigations, however, the radiochemist is frequently confronted with the task of preparing the pure substance. But it will be clear from what has been said that in this case he must be informed exactly as to the day and hour at which the 'radioactively pure' preparation is desired, and he will then so arrange his work that the short-lived and hence quickly regenerated disintegration products are removed immediately before the preparation is to be used.

A second difficulty met with in the preparation of pure radioactive substances lies in the fact that many, in fact most, of the radioactive substances cannot be separated chemically from other partly active and partly inactive substances, once they have been mixed, either during their preparation or in consequence of their occurrence together in nature. This difficulty is a much more disagreeable one, because it is capable of making impossible the practical utilization of the radioactive substances. These disturbing elements are isotopes of the substance concerned (see Chap. XV). Radium D, for example, in spite of the relatively large quantity of it which occurs in minerals, and although its chemical properties are quite accurately known, cannot be obtained in concentrated form, in which form it would be very useful for many purposes, because its chemical properties are identical with those of the other types of lead with which it is associated in minerals. We now know that the phenomenon of isotopy is one which also occurs with numerous inactive elements, but in this case the problem of the separation of the isotopes manifests itself in quite another form. Here we require to separate the old chemical elements into fractions differing from each other solely in atomic weight, the solution of which task is sought mainly because of its theoretical importance. But with the radioelements the individual atomic types of the elements are characterized by especially interesting and valuable properties, their radiations, and hence the problem of the separation of isotopes appears here as the preparation in the 'pure state' of the radioactive substance concerned.

A final difficulty met with in the isolation of the radio-elements has its origin in the short life of some of them. When they decay in fractions of a second, it is clear that we cannot obtain them by chemical manipulations such as precipitation, filtration, and so on; but even those which live for several minutes or hours cannot generally be isolated chemically from such a complicated mixture as that presented by most uranium and thorium minerals. This difficulty can be avoided, however, for the quicker an element decays the more rapidly it will be regenerated from its parent substance. Correspondingly, the general principle involved in the preparation of short-lived substances consists in separating the parent substance from the mineral in as concentrated a form as possible. Should this parent substance also be too unstable, we should separate an earlier more stable member of the disintegration series. We can then await the growth of the desired product, and undertake the chemical separation in a manner which is generally both easy and quick. Table 45 gives a summary of the radio-elements which, in consequence of their long half-value periods (T), entered in the table, can be extracted directly from the minerals. They are arranged in the table in the order of decreasing half-value period, and the last five can also be obtained from their parent substances, just as in the case of the typically 'short-lived' elements. Under favourable circumstances this is a particularly easy process with RaD, for one of its more distant ancestors, radon, is easily prepared in the pure state, and the intermediate products are all short-lived.

TABLE 45

Radio-elements of Long Life which can be extracted directly from Minerals

Thorium	$T = 1{\cdot}34 \times 10^{10}$ years	Radium D†	$T = 22$ years
Uranium	$4{\cdot}4 \times 10^{9}$ years	Actinium	13·5 years
Ionium†	$8{\cdot}3 \times 10^{4}$ years	Mesothorium 1†	6·7 years
Protoactinium	$3{\cdot}2 \times 10^{4}$ years	Radiothorium†	1·9 years
Radium	1,590 years	Polonium	140 days

Those elements denoted by † cannot be extracted directly as pure elements owing to the fact that the minerals contain isotopes of longer life.

In what follows we shall give separate accounts of the most important facts concerning the preparation of the long-lived and of the short-lived radio-elements.

A. PREPARATION OF THE RADIO-ELEMENTS OF LONG LIFE

Thorium. Since large quantities of very pure thorium are required in industry, quite a number of processes for the extraction of 'chemically pure' thorium are known. But we shall not deal with them here, because fuller information can be obtained from text-books on chemistry and particularly from special works on the rare earths. It should be mentioned here, however, that the thorium derived by the usual processes is never 'radioactively pure', but is contaminated at least by its isotope radiothorium. Nevertheless, there is a means of separating thorium even from this radio-element, and we propose to give it, since it is instructive to see how the chemist, by taking time into his confidence, can carry out purifications that would be impossible by direct means. The radiothorium contained in thorium after its chemical purification decays to a negligible fraction of its former value after the lapse of 20 years; but during this time mesothorium 1 is formed from the thorium, and from this fresh radiothorium is produced. By adding barium to the solution and separating the thorium from it, we can, by frequent repetition of this process throughout the twenty years, ensure that appreciable quantities of mesothorium can never accumulate in the thorium solution. During this period the old radiothorium has completely disappeared and no fresh radiothorium has been regenerated in the solution, so that we now have not only the chemically pure thorium salt in solution, but it is also pure from the radioactive point of view. In order that this may be successful, however, it is necessary that the thorium mineral should contain only very small amounts of uranium, so that the separated thorium will be contaminated with the merest traces of the long-lived isotope ionium.

Monazite sand is used almost exclusively as the source for the extraction of thorium. The best-known thorium minerals are thorite and thorianite (see also Table 52, p. 273).

Uranium. We do not propose to deal with the question of the preparation of chemically pure uranium salts either, as the necessary instructions are readily found in the literature of chemistry. In contrast to thorium, we cannot obtain uranium as a 'pure element' no matter what artifice we may adopt. Since actino-uranium and uranium II, which are isotopic with uranium I ($T = 4{\cdot}4 \times 10^9$ years), possess large half-value periods, $T = 7 \times 10^8$ and 3×10^5 years respectively, it is impossible to wait till they have decayed. But for all practical purposes (i.e. as regards the magnitude of its combining weight, etc.) the 'mixed element' uranium consists only of uranium I; the amount of AcU is less than 1 per cent., and that of U_{II} less than one-hundredth per cent. of the amount of U_I.

Pitchblende is the chief mineral now used in the manufacture of uranium (see Table 52, p. 273).

Radium. In all unaltered primary uranium minerals about $3{\cdot}4 \times 10^{-7}$ gm. of radium is associated with 1 gm. of uranium. Correspondingly, we find only about 2 gm. of radium in 10,000 kg. of a uranium pitchblende. Its extraction is a very tedious task, of course, but it is not disturbed by the presence of any isotope of radium, and it is due to this happy circumstance that the first attempt to extract radium from pitchblende residues was crowned with complete success. This work was carried out by M. and Mme Pierre Curie. Since the chemical properties of radium are very closely related to those of barium, we can apply any process which permits a quantitative and rapid separation of barium from the remaining constituents of the mineral. It is not possible to give a definite and generally valid procedure for the extraction of radium, because the initial materials require different kinds of treatment. Moreover, the price of radium is not high enough to justify the rejection of the other valuable constituents of the minerals, without endangering the business aspect of the process. Thus the uranium content is about one-third as valuable as the radium content, and when vanadium is present it must always be extracted. Nevertheless, in all the processes, we can differentiate roughly between the following main stages in the extraction:

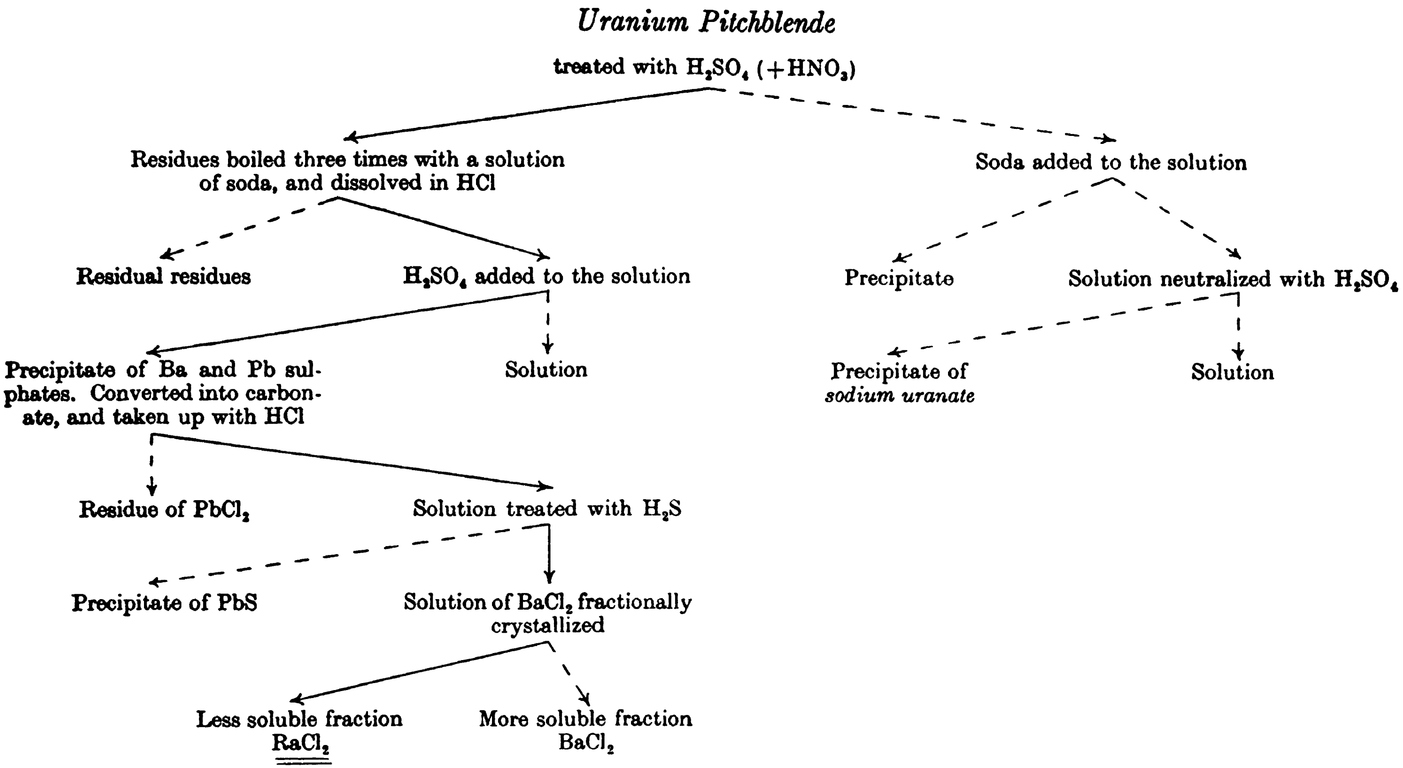
1. Solution or opening up of the uranium minerals.
2. Separation of all sulphates insoluble in water.
3. Their conversion into soluble salts.
4. Purification of the radium and barium from foreign constituents.
5. Fractional enrichment of the radium in the barium.

Each of these points can be widely varied, and we do not propose to enter into the numerous suggestions and experiments on this aspect of the subject. We shall content ourselves with the discussion of the main features of two processes that have been well tried on a large scale, and which differ greatly both in the initial materials used and in the methods adopted for their treatment.

(a) *Process in Use at St. Joachimstal (Jáchymòv)*

St. Joachimstal in the Erzgebirge is the most important source of radium-bearing ores in Europe. Here the process described below has been applied continuously since 1910, originally under the control of the Austrian Government, and since the mines and factory were taken over by the Czechoslovak authorities, nothing has been altered in the methods originally introduced.

The initial material is uranium pitchblende, the main constituent of which is U_3O_8, with admixture of very many other compounds, especially PbS, SiO_2, CaO, FeO, and MgO. It contains crystalline material in a highly dispersed state, the size of the individual crystals being only 10^{-4} to 10^{-7} cm. The broken-up ore is treated with sulphuric acid to which some nitric acid has been added, the concentration being such that the uranium goes completely into solution, whereas the radium remains behind quantitatively with the insoluble sulphates of calcium, barium, and lead. These are more or less completely taken up by boiling three times with a solution of soda, the parts which still remain unchanged, the so-called 'residual residues', containing among other things the whole of the tantalum, and to these we shall return in


Uranium Pitchblende
treated with H_2SO_4 (+HNO_3)
Residues boiled three times with a solution of soda, and dissolved in HCl
Soda added to the solution
Residual residues
H_2SO_4 added to the solution
Precipitate
Solution neutralized with H_2SO_4
Precipitate of Ba and Pb sulphates. Converted into carbonate, and taken up with HCl
Solution
Precipitate of sodium uranate
Solution
Residue of $PbCl_2$
Solution treated with H_2S
Precipitate of PbS
Solution of $BaCl_2$ fractionally crystallized
Less soluble fraction $RaCl_2$
More soluble fraction $BaCl_2$

connexion with the extraction of protoactinium. For the purpose of purification the dissolved constituents are again precipitated as sulphate, converted into carbonates and then into chlorides. In the process the lead remains partly undissolved as chloride, and it is partly removed by means of precipitation with sulphuretted hydrogen. The final part of the process is the fractional crystallization of the mixture of radium and barium chlorides until the required concentration of the radium has been attained. Owing to its smaller solubility, the radium is concentrated in the crystals.

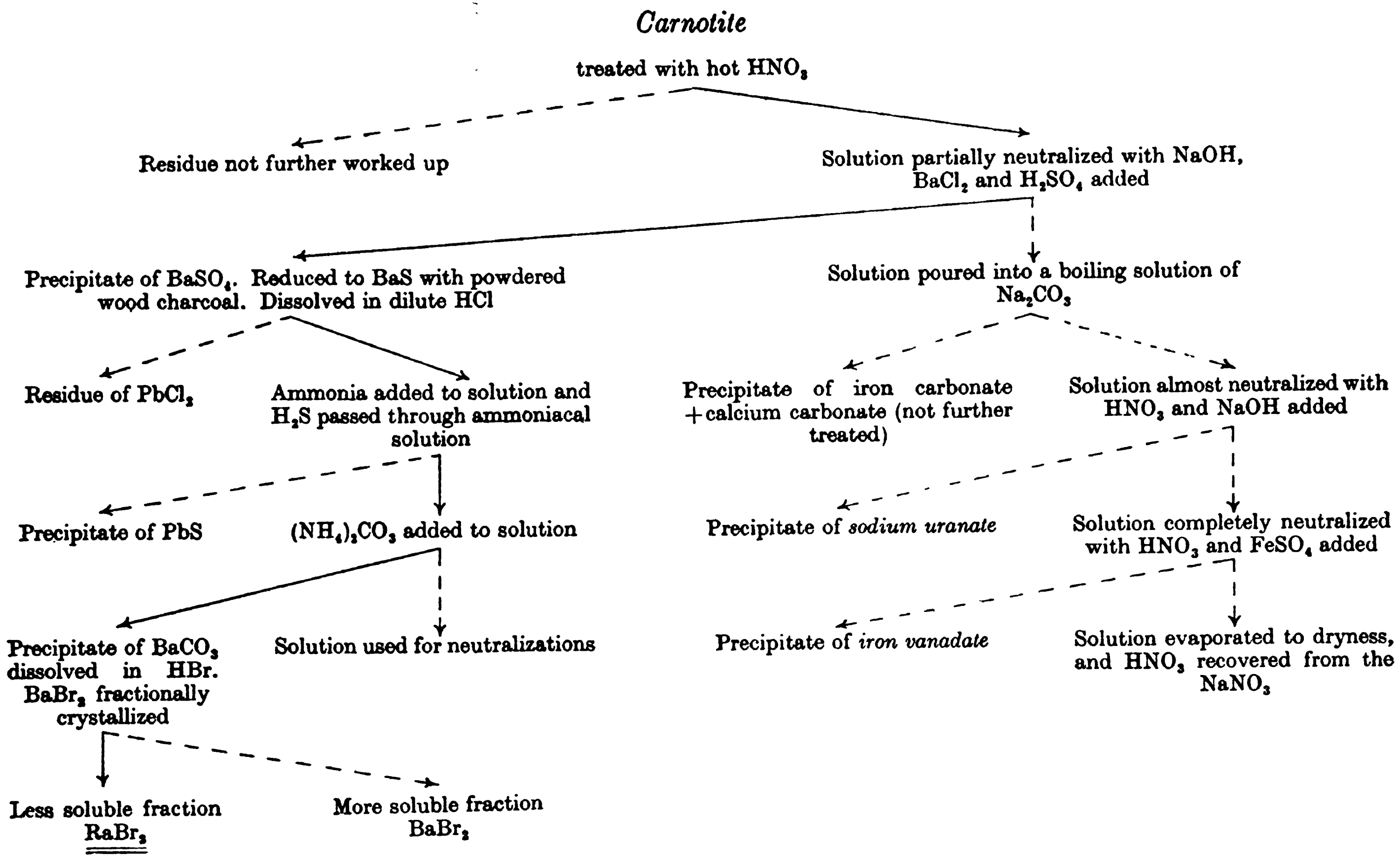
In order to make the various steps in the process of extraction as clear as possible a schematic representation of the process is given on p. 208.

The quantity of radium produced per annum at St. Joachimstal only amounts to 3 or 4 gm., owing to the difficulties to be overcome in the mining of the uranium ore. During recent years a radium factory has been worked at Oolen (near Antwerp) in Belgium, and the annual production of radium is here very much greater than that of St. Joachimstal. The mineral used in the production of radium in the Belgian factory is a rich pitchblende which occurs at Katanga in the Belgian Congo. As the initial material is pitchblende, the essential features of the method of treating the mineral will not differ from those in use at Joachimstal, but details of the process have never been published. The yearly production of radium in Belgium at present amounts to about 36 gm.

(b) *Process in Use at Denver*

As a second illustration of the rational extraction of radium, we shall now describe the process developed in the years 1914 and 1915 at Denver in the State of Colorado, North America. The deviations from the Austrian process are due mainly to the fact that the initial material is carnotite, a vanadate of potassium and uranium of composition $K_2O \cdot 2UO_3 \cdot V_2O_5 \cdot 2H_2O$, but also partly to the fact that the resources available for the equipment of the plant were much greater than in the previous case.

The ore is treated with hot concentrated nitric acid to which a small amount of hydrochloric acid has been added, and the whole of the radium is thereby brought into solution, in spite of the presence of sulphates. After partial neutralization with NaOH and addition of barium, it is then precipitated as sulphate together with barium and lead. The decanted solution is worked up for uranium and vanadium, whilst the sulphates obtained are mixed with wood charcoal in a graphite crucible, heated for 7 hours at 800° C., and so converted into sulphides. These are dissolved in hydrochloric acid, the lead is removed by precipitation with hydrogen sulphide in ammoniacal solution, whereas the


Carnotite
treated with hot HNO_3
Residue not further worked up
Solution partially neutralized with NaOH, $BaCl_2$ and H_2SO_4 added
Precipitate of $BaSO_4$. Reduced to BaS with powdered wood charcoal. Dissolved in dilute HCl
Solution poured into a boiling solution of Na_2CO_3
Residue of $PbCl_2$
Ammonia added to solution and H_2S passed through ammoniacal solution
Precipitate of iron carbonate + calcium carbonate (not further treated)
Solution almost neutralized with HNO_3 and NaOH added
Precipitate of PbS
$(NH_4)_2CO_3$ added to solution
Precipitate of *sodium uranate*
Solution completely neutralized with HNO_3 and $FeSO_4$ added
Precipitate of $BaCO_3$ dissolved in HBr. $BaBr_2$ fractionally crystallized
Solution used for neutralizations
Precipitate of *iron vanadate*
Solution evaporated to dryness, and HNO_3 recovered from the $NaNO_3$
Less soluble fraction $RaBr_2$
More soluble fraction $BaBr_2$

barium and radium are precipitated as carbonates, and again dissolved in hydrobromic acid. In this case the enrichment of the radium in the barium is carried out by fractional crystallization of the bromides.

The Denver process is represented schematically on p. 210, the operations leading to the extraction of uranium and vanadium being included also. The American carnotites contain on the average 4 mg. radium per metric ton. Up to the end of 1922 about 52,000 tons of the ore had been worked up, corresponding to the production of about 200 gm. of radium. Since the opening of the Belgian factory (see above) both the American and Joachimstal productions of radium have lost in importance year by year, and from time to time the factories have been closed down entirely. At the same time the price of radium has gone down (see p. 224).

The virtual monopoly held by the Belgian producers of radium for more than a decade has recently come to an end. In 1930 an extensive occurrence of pitchblende was discovered on the eastern shores of the Great Bear Lake, North West Territory, Canada [1]. This pitchblende resembles the Joachimstal deposits in its general character and in the nature of the metallic minerals associated with it. Picked samples of the ore contain from 45 to 55 per cent. of U_3O_8, and the thorium content of the ore is almost negligible (*c.* 0·004 per cent.). Lead extracted from it is found to be fairly pure RaG, for the combining weight of this lead is 206·06, and the mineral contains round 10 per cent. of lead. As the most northerly point reached by the railway system is over 1,000 miles distant from the Great Bear Lake, the transport of the material has presented very great difficulties. These have been surmounted by the conveyance of the concentrated ores by boat during the short period when rivers and lakes are not ice-bound, and during the rest of the year they are transported by air to the nearest railway terminus. A radium refinery has been erected at Port Hope, Ontario, and already 3 gm. of radium are being produced monthly. It is hoped that the output will soon be doubled. The undertaking is working smoothly, and in all probability it will effect a further reduction in the international price of radium (see p. 224). Finally, it may be mentioned that, during the past few years, the Chemical Works at Treibach in Carinthia (Austria), working on ores from various sources, have been able to produce several grams of radium annually.

From the discovery of radium to the end of 1936 it is estimated [2] that the amounts of radium produced from ores derived from various sources are as follows:

St. Joachimstal (Jáchymòv) .	50 gm.	Canada (Great Bear Lake) .	28 gm.
United States of America .	250 ,,	Portugal, Russia, England, etc.	50 ,,
Belgian Congo (Katanga) .	500 ,,	World production	= *c.* 900 gm.

Mesothorium 1. We shall proceed immediately to the description of the extraction of mesothorium, because this element is isotopic with radium, and hence the extraction of the two substances is carried out by very similar processes. Mesothorium is found in every thorium mineral, there being $3{\cdot}7 \times 10^{-10}$ gm. present with each gram of thorium. But since all thorium minerals also contain a small quantity of uranium, and 1 gm. of uranium corresponds to the presence of $3{\cdot}4 \times 10^{-7}$ gm. of radium, it follows that all mesothorium extracted from minerals must be mixed with an appreciable quantity by weight of radium, even though the radiation from it may be much weaker. Mesothorium can be obtained as a 'pure element' only by awaiting its gradual regeneration in thorium salts. It can be isolated simply by precipitation of the thorium with ammonia free from carbonates, eventually after the addition of a little barium, from which the mesothorium can be separated again later by fractional crystallization. But it is self-evident that, even after the lapse of years, only a very small yield by weight can be obtained in this manner. In all practical applications, the radiation value of a mesothorium preparation is increased if it contains any of the longer-lived element radium.

The mesothorium content of radium is generally very small. If the radium has been derived from American carnotite, about $1{\cdot}5 \times 10^{-3}$ of the γ-radiation is due to mesothorium plus radiothorium, whereas if it was obtained from Joachimstal pitchblende this fraction only amounts to 6×10^{-6}.

The extraction of mesothorium on a large scale is carried out in conjunction with that of thorium. In the working up of monazite sand a residue insoluble in sulphuric acid is obtained, and these insoluble sulphates are then worked up in exactly the same manner as that which has been described for the extraction of radium. If there is any doubt as to whether the initial material contains sufficient barium, it is advisable to add a little of a barium salt.

Protoactinium. In the extraction of radium by the Joachimstal process it was mentioned that the 'residual residues' were those parts of the initial material most difficult to bring into solution, and that in addition to acids of the rare earths they also contain tantalum and its higher homologue protoactinium. We obtain a fraction of similar composition by working up pitchblende with sodium bisulphate. By treatment with hydrofluoric acid the acids of the rare earths can be brought into solution, whereas barium and lead remain undissolved. On subsequently digesting with sulphuric acid we again obtain a deposit of tantalic acid, which can be freed from iron and other impurities by means of aqua regia. Another method consists in dissolving out the tantalum directly from the pitchblende by means of hydrofluoric acid and sulphuric acid. Protoactinium is markedly more basic than tantalum and shows a very close relationship to zirconium and hafnium. In the preparation of pure protoactinium use is made of this relationship, the protoactinium being precipitated with zirconium phosphate. The second step consists in the separation of the phosphoric acid by means of alkalis, and the conversion of the zirconium and protoactinium into oxychloride. The separation of zirconium is carried out by means of oxalic acid, after addition of thorium. Finally, the protoactinium is separated from the thorium by the use of hydrofluoric acid. Since the amount of uranium mineral required for the preparation of 1 gm. of radium contains over 0·5 gm. of protoactinium, it has been possible to prepare the latter in a pure state in quantities of this order of magnitude [3].

Actinium. This radio-element is a higher homologue of lanthanum and can be extracted together with this element from all uranium minerals, by means of the methods of separating rare earths dealt with in treatises on chemistry. The concentration of the actinium as compared with the lanthanum salts is, however, met by difficulties similar to those which generally arise in the separation of the rare earths from one another; but the difficulties are intensified on account of the fact that the radiological control of concentration is very tedious, owing to the necessity for awaiting the development of disintegration products from the rayless† actinium. Hitherto it has been established that actinium is more strongly basic than lanthanum, and it is therefore only incompletely precipitated by ammonia, particularly in the presence of ammonium salts. A more convenient method of obtaining it will be rendered accessible when a sufficient amount of its parent protoactinium has been prepared in the pure state; owing to the difference of their chemical properties it will be a simple matter to separate the actinium from the protoactinium.

Ionium. Up to the present we know of no uranium mineral that is absolutely free from ordinary thorium, and for this reason it has not yet been possible to prepare the isotopic element ionium in the pure state. But the mixed element obtained from uranium minerals practically free from thorium consists of equal parts of ionium and thorium, and it accordingly exhibits an appreciably smaller combining weight (see p. 151). Conversely, every thorium mineral contains appreciable quantities of uranium from which slight traces of ionium are formed, and hence, although the atomic weight of the thorium obtained is not altered thereby, its activity is always more or less increased by the presence of this admixed ionium. It is clear that, for the separation

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† The β-radiation from actinium, the existence of which it is necessary to assume in order to explain its transformation into radioactinium (cf. p. 229), is too weak to be detected.

of thorium-ionium salts from minerals, the methods that have been developed for ordinary thorium are applicable.

Regarding *radiothorium* it has already been mentioned that it is always obtained together with thorium from minerals. It can be prepared as a pure element if we start off with mesothorium in a state of radioactive purity; radiothorium is gradually generated in the mesothorium, and after the lapse of sufficient time a simple barium-thorium separation can be carried out by means of NH_3. Since the regenerated radiothorium is only present in unweighably small quantities, it is necessary, in order to obtain visible precipitation, to add, say, a ferric salt which is precipitated with ammonia. In this connexion reference should be made to the chemical treatment of the radio-elements of short life (p. 215).

Radium D. Although RaD is itself almost rayless, it is the constituent of radio-lead which is responsible for the activity of several decades duration, inasmuch as it is the parent of radium E and polonium. It can never be obtained in the pure state from minerals, for even though there may be no ordinary lead in the uranium mineral concerned, the latter inevitably contains radium G, which is likewise an isotope of lead. Moreover, as compared with the amount of RaG present, the amount of RaD in the quantitative sense is negligible. On the other hand, RaD can be obtained as a pure element from radon, although the amount obtainable in this way is very small. But since this method belongs to those which can only be carried out in general with short-lived radio-elements, we shall defer the discussion of it till later. It is worth mentioning, however, that RaD associated with ordinary lead is also a very valuable material for radiologists, for it serves as a useful source from which RaE and polonium may be periodically obtained.

Polonium. This element can either be separated together with bismuth directly from uranium minerals, or it can be obtained more conveniently from radio-lead. As already mentioned, polonium is being generated continuously in this mixture of ordinary lead with its isotopes RaD and RaG, until it is in equilibrium with the RaD present. Moreover, it is possible to obtain it in fairly concentrated form by recrystallizing the radio-lead, preferably in the form of nitrate. The polonium remains in the mother liquor, and may finally be obtained electrolytically and free from lead and RaE by maintaining a cathode potential that is not allowed to fall below $E_c = -0{\cdot}08$ volt.† Gold or molybdenum is to be recommended in preference to platinum as the

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† Since it is rather troublesome to measure the individual potential of the electrode, we may make use of the following approximate instructions, which are quite satisfactory in practice. If the current be not allowed to exceed 3×10^{-5} amp. per sq. cm. of the cathode surface, the nitric acid solution being $\frac{1}{10}$ N in strength, the polonium is extracted in a very pure state.

material of the electrodes, because the polonium cannot be redissolved quantitatively from the last-named metal, owing to the formation of an alloy. (Further details on the electrochemical separation of polonium are given in Section B, in connexion with the preparation of RaA, RaE, ThB, and ThC.)

B. PREPARATION OF THE RADIO-ELEMENTS OF SHORT LIFE

The shorter the life of the radio-element, the more necessary it is for the methods of preparing and purifying it to differ from the ordinary chemical ones, and the more necessary it also becomes to have recourse to measurements of activity as a means of controlling these methods. In addition, the radio-elements of short life occur for the most part towards the end of the disintegration series, later on than the emanations. This circumstance also makes it advisable to describe them separately, for we shall see that the ease with which the emanations can be obtained in the pure state can also be utilized in the isolation of their succeeding products. For this reason we shall consider first the methods of preparing the three emanations.

Being inert gases, the *emanations* are only occluded and never chemically attached to the salts in which they are formed. They usually escape partially even at room temperature, whilst the major portion is liberated by heating, and if the salt concerned be fused, the whole of the emanation can be set free. By carrying out the fusion in a suitably designed apparatus the whole of the emanation can be collected. The hydroxides and halides 'emanate' best, even at room temperatures, whereas sulphates retain large quantities of the emanation. In order to collect radon (radium emanation) quantitatively, it is most convenient to dissolve radium chloride or bromide in water, to allow the sealed-off solution to stand until a sufficient quantity of emanation has been generated, and then to pump off the whole of the gases and transfer them to another vessel. By suitably constructing this vessel we may use it to perform the separation of the rare gases from the admixed gases nitrogen, oxygen, hydrogen, and ozone, by the methods used in gas analysis. The oxygen, hydrogen, and ozone are always formed in consequence of the action of the rays from radium on the water (see Chap. XXIV). It often suffices to explode the explosive mixture of hydrogen and oxygen, which constitutes the major portion of the mixed gases, by sparking in a eudiometer tube similar to those used in gas analysis. If it is required to obtain the radon in still purer form, it is advisable next to free the residual gases from carbon dioxide and water vapour by contact with solid potassium hydroxide, and then to condense the radon by means of liquid air. The uncondensed impurities (nitrogen, hydrogen, rare gases) are then pumped off, and after removal of the liquid air the regasified radon is allowed to stream into the vessel in which it is to be used.

It is obvious that such a complicated purification can only be carried out with the relatively long-lived emanation radon; in the cases of thoron and actinon we have to content ourselves by liberating and collecting them from the salts or solutions, even though they are contaminated by large quantities of inactive gases. They can never be utilized in the form of such intense sources of radiation as radon, but for most purposes, especially the extraction of their disintegration products, we are not disturbed by the presence of the admixed inactive gases.

The first disintegration products of the emanations are the 'short-lived active deposits', this being the group-name for the products radium A, radium B, radium C, radium C′, radium C″, and the corresponding disintegration products of the thorium and actinium disintegration series. They are usually obtained by hanging a negatively charged platinum foil in the vessel containing the emanations. The yield is greater the larger the surface of the platinum foil, and its negative charge only serves as a means of concentrating the active deposit upon it. The above-named products are deposited on the foil, and can be obtained in solution by means of dilute acids, and in large measure even with hot water. If necessary they can then be separated. In order to obtain thorium B and thorium C it is very convenient to use an apparatus like that depicted in Fig. 42 (p. 133); situated at the bottom of this apparatus is a glass or platinum dish containing the salt of radiothorium or thorium X.

By a shorter or longer exposure of the platinum foil in the emanation we have a means of altering in a known manner the relative proportions of the A-, B-, and C-products, so as to get a favourable yield of the substance desired. In this way it is not difficult by means of very short exposures to obtain practically pure *radium A* (cf. p. 135). As a rule, however, it is necessary to effect a subsequent separation of the individual members of the active deposit. In the thorium and actinium disintegration series the A-products hardly call for consideration, owing to their very short life, and even radium A decays so rapidly that its removal presents no difficulty, if we require only radium B plus radium C. It suffices to wait about 20 minutes after completion of the exposure before using the active deposit. We have just described the method of obtaining pure radium A, so that we need now only discuss the methods for the preparation of the *B-* and *C-products* in the pure form.

We can achieve a partial separation simply by heating the platinum foil; thus at 800° C. 75 per cent. of thorium B volatilizes and only

20 per cent. of thorium C. But the chemical and electrochemical methods of separation are more effective.

In order to apply them we first proceed to dissolve the active deposit from the platinum foil by means of dilute acids, and then make use of the known methods of separating bismuth and lead. Thus, by means of electrolysis, if the cathode potential reaches the approximate value of $E_c = -0{\cdot}5$ volt, but does not fall below this limit—in a weakly acid solution this corresponds to a current density of 0·4 ma. per sq. cm.—we can separate pure C, whereas the B remains almost quantitatively in solution. The separation of B can be almost completely prevented if we add to the solution a small quantity of isotopic lead in the form of a soluble lead salt, and so ensure that the very small quantity of lead that is separated out below the above-mentioned cathode potential, which is characteristic of lead in this state of dilution, consists almost wholly of inactive lead atoms. If the whole of the bismuth isotopes have been removed, the deposition of which already begins at a potential of $E_c = -0{\cdot}08$ volt, corresponding to about 0·16 ma. per sq. cm., it suffices to increase the terminal voltage so that the cathode potential is below $-0{\cdot}5$ volt in order to be able to separate out the B-products on an arbitrary electrode. If these are required, we should of course avoid previously adding appreciable quantities of inactive lead.

Instead of carrying out an electrolytic separation, a still simpler method of separating out the C-products is to dip a clean sheet of nickel into a hydrochloric acid solution of B and C. As a result of a process which is essentially identical with that of electrolysis, the C-product is deposited on the nickel sheet free from the B-product (cf. p. 162). If platinum be modified electrochemically by charging it with hydrogen, polonium, RaE, ThB, and ThC are deposited on it, without the passage of a current [4].

It is never possible by means of acids to effect the quantitative solution from metal sheets of the radio-elements or of their oxides, which are always present when the sheets are exposed to air. This is due to the occurrence of diffusion of the radio-elements from the surface to the inside of the metal. When this diffusion takes place very slowly, as with polonium on gold, it is possible to effect fairly complete solution of the radio-element from the sheet by means of acid (see p. 215).

It should be emphasized that *radium E*, since it is isotopic with bismuth, can be obtained electrochemically by the same methods as apply to the C-products, and that *radium A* and its isotope *polonium* are still more readily deposited, even at a cathode potential of $E_c = -0{\cdot}35$ volt. In this case we can increase the degree of purity of the radium A or polonium by adding a salt of bismuth, when the deposition of radium C or radium E along with the radium A or polonium is rendered impossible. If it is necessary to separate radium E and polonium from radio-lead, it is advisable to add antimony, and to precipitate with pyrogallol in weakly acid solution; the radium E and polonium are carried down with the antimony, and can then be separated from it electrolytically in the presence of tartaric acid.

In the electrochemical separation of polonium and radium E, radio-lead from minerals is not the only source from which they may be derived. We may also use the 'active deposit of long life', which is formed when we allow radon to decay in a closed vessel. *Radium D* is then found on the walls of the vessel, and can be removed by solution in acids. Radium E and radium F (polonium) are generated continuously in this solution, and owing to their greater concentration they can be obtained more conveniently from it by one of the above-described electrochemical processes than from radio-lead, which is soluble only in larger quantities of liquid. The best method of proceeding is first to make the solution $\frac{1}{2}$ N with hydrochloric acid and to move about in it a small clean sheet of silver, which effects practically a quantitative separation of the polonium. The solution is then evaporated to dryness, taken up with a 0·2 per cent. solution of hydrochloric acid, and the RaE is deposited on a small clean sheet of nickel. Finally, if we wish to electrolyse out also the radium D, say for the purpose of its purification, it is advisable to separate it anodically as lead peroxide, which is readily accomplished in a 7 per cent. solution of nitric acid with a current density of about 3×10^{-4} amp. per sq. cm., using platinum electrodes. It may be mentioned that the device of depositing the radio-element anodically as peroxide is often suitable also for the purification of polonium.

For radio-elements to which they can be applied the electrochemical methods of separation are generally preferable to all others, mainly because in this way the substances are obtained in a state of chemical as well as of radiochemical purity. Unfortunately they can only be contemplated for those radio-elements which are to some extent electrochemically noble, and hence especially for the isotopes of lead, bismuth, and polonium. In the preparation of very strong polonium sources the electrochemically separated polonium, deposited preferably on rotating electrodes [5], is later concentrated by a process of volatilization and condensation on a smaller surface.

Short-lived thorium isotopes like uranium X_1, radiothorium, and others can be deposited electrochemically on platinum wires from a hydrochloric acid solution, with a yield of about 60 per cent.

Another very elegant method of separation is based on the utilization of the phenomenon of 'radioactive recoil' (see Chap. VI), but its range of applicability is still more restricted. We may with advantage obtain the thallium isotopes *thorium C″* and *actinium C″*, when the particles of these substances recoil in air from a metal plate coated with the active deposit, by collecting them on a negatively charged metal place placed opposite to the activated metal plate. We may also derive pure *radium B* in this way by recoil from radium A, but in this case we must terminate the experiment after the lapse of a few seconds, for otherwise the radium B collected will be mixed with radium C that has been generated from it.

In conclusion, we must deal with those methods of preparing and separating short-lived radio-elements which are most generally applicable, the *chemical methods*. We have accurate information on the chemical nature of all the radio-elements of short life. We know that we are dealing with different kinds of thallium, lead, bismuth, and polonium—these are the products A to G; further, that thorium X and actinium X are isotopes of radium, whereas uranium X_1, uranium Y, and radioactinium are isotopes of thorium, and that mesothorium 2 is identical chemically with actinium, and uranium X_2 with protoactinium. Hence the analytical chemistry of these substances can only afford difficulty inasmuch as they are always present only in exceedingly small concentration. This will be clear if we recall that in ordinary radiochemical work a content of 10^{-11} mol of thorium B per litre would be pronounced a fairly 'strong' solution. An artifice which must always be successful in overcoming the difficulties attendant upon the extreme dilution of these substances consists in the addition of weighable quantities of an isotope. Thus, if we wish to remove thorium B quantitatively from a solution, it suffices to add a little lead nitrate and precipitate the entire lead by means of sulphuretted hydrogen. All the atoms of the mixed element lead so obtained, and consisting of ordinary lead and thorium B, are chemically identical, so that by passing sulphuretted hydrogen through the solution the thorium B must be removed to the same high degree as the lead in the form of practically insoluble lead sulphide.

A disadvantage of this method consists in the fact that once such mixing has taken place the process is practically irreversible, and hence the radio-element can never again be obtained in concentrated form. For this reason we should only revert to this method of adding an isotope when we are concerned with the quantitative removal and not with the preparation of a radio-element. In the latter case it is always advisable not to use an isotope but to have recourse to a related element as a carrier in the precipitation reaction. This is analogous to the addition of barium in the extraction of radium or mesothorium already discussed, for in this way we can effect a separation from the more distant chemical elements, and still finally bring about the separation of the radio-element from the added barium. Similarly, it is often better to precipitate, say, radium E by the admixture of lead rather than of bismuth, for then it can easily be separated again by electrochemical means (see above). Likewise we should make use of zirconium or cerium rather than thorium for the separation of uranium X_1, and so on.

When a related element and not an isotope is used, we can no longer maintain that the radio-element must also be removed by every quantitative precipitation of this substance. Lead sulphate, for instance, would leave radium E in solution, whereas lead sulphide would remove it quantitatively. In such cases the adsorption and precipitation rule which we have already discussed is applicable (see p. 163 et seq.). A radio-element will be adsorbed and precipitated by those deposits the anions of which form with the radio-element concerned a compound which has a low solubility in the solvent used. In the place of the isotope we should thus be able to use any other element for mixing purposes, so long as it forms, like the radio-element, an insoluble precipitate with the anion in the solution. Instead of the hydroxide precipitation of the thorium isotopes we may utilize the hydroxide precipitation of iron salts, or in place of the sulphate precipitation of the lead isotopes we may use the sulphate precipitation of barium salts, and so on.

It is not necessary for us to do more than to direct attention to these rules here, since they have done invaluable service in the entire large domain of the analytical chemistry of the radio-elements, in spite of a few exceptions that have not yet been fully explained. Our knowledge of these rules enables us to give a clear summary of the methods of separation of the radio-elements of short life which have been tested hitherto, and to predict new and suitable methods. It must be left to the analytical experience of the chemist to decide which individual process is to be recommended, in accordance with the special problem on hand.

REFERENCES

For further information on the topics of this chapter reference should be made to *Le Radium et les Radio-éléments*, by Maurice Curie (Baillière, Paris, 1925), where the development of the radium industry up to 1925 is fully treated, to *Radium und Isotope*, by O. Erbacher (Gmelin's *Handbuch der Anorgan. Chemie*, 8th edn., Berlin, 1928), and to the books mentioned at the end of Chap. XV (p. 153).

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XXIII

THE PROPERTIES OF THE INDIVIDUAL RADIO-ELEMENTS

IN this chapter we shall discuss the individual radio-elements in the order of their production from the parent substances uranium, protoactinium, and thorium, and in each case we shall give the most important data which serve to characterize that radio-element from the physical and chemical points of view. It will suffice if we are brief here, because in the preceding and more general chapters we have also frequently made reference to the special properties of the radio-elements. Thus we shall only deal with the methods of preparation in so far as in one case or another the systematic treatment given in Chapter XXII requires to be supplemented from the practical viewpoint. In conclusion, we shall devote some space to the radioactive elements potassium, rubidium, and samarium, which stand in an isolated position as compared with the rest of the radio-elements, and which have not yet been so thoroughly investigated as these.

The significance of the symbols used is as follows: Z = atomic number; A.W. = atomic weight; C.W. = combining weight; T = half-value period†; R = range in cm. in air at 760 mm. pressure and 15° C.; μ = coefficient of absorption in aluminium, measured in cm.^{-1}

A. THE URANIUM SERIES

1. Uranium

Uranium is a mixture of three isotopes, uranium I (^{238}U), uranium II (^{234}U), and ^{235}U, which is probably identical with actino-uranium, the parent of the actinium series [1] (see also p. 230). The period of uranium II being so much shorter than that of uranium I, the mixed element 'uranium' contains only 0·008 per cent. of uranium II, and less than 1 per cent. of the isotope ^{235}U. The rate of decay of ^{235}U is not exactly known, but it is probably about 6 times that of

	Uranium I	*Uranium II*
Z	92	92
A.W. . . .	238·07	234
T	$4{\cdot}4 \times 10^9$ years	$3{\cdot}4 \times 10^5$ years
Radiation . .	α	α
R	2·63 [2]	3·18 [2]
Parent element .	—	UX_2
Daughter element .	UX_1	Io

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† The disintegration constant (λ) can be calculated simply from T by means of the relation $\lambda = \frac{\log_e 2}{T} = \frac{0{\cdot}693}{T}$ (see p. 127).

uranium I. We shall not deal with the chemical properties of uranium here, as they are to be found in every text-book of chemistry. In radioactive investigations we generally use the grey-black oxide of uranium U_3O_8 or the yellowish fluorescent uranyl nitrate $UO_2(NO_3)_2 \cdot 6H_2O$, the former as a standard for α-ray measurements, and the latter for the preparation of uranium X. For information as to the occurrence of uranium minerals see Table 52, p. 273, and on methods of extracting radium from them see p. 207 et seq.

The α-activity of uranium in equilibrium with all its disintegration products is 4·73 times as large as that of uranium alone. The total α-radiation from 1 gm. of uranium in the form of an infinitely thin layer, and utilizing both sides of the layer, would maintain in an ionization chamber a current of 1·37 e.s.u., or $4{\cdot}56 \times 10^{-10}$ amp. A layer of U_3O_8 of area 1 cm.^{2}, if only the radiation from one side of it be taken into account, would maintain in an ionization chamber a current of $1{\cdot}73 \times 10^{-3}$ e.s.u., or $5{\cdot}76 \times 10^{-13}$ amp.

2. Uranium X_1 and Uranium X_2

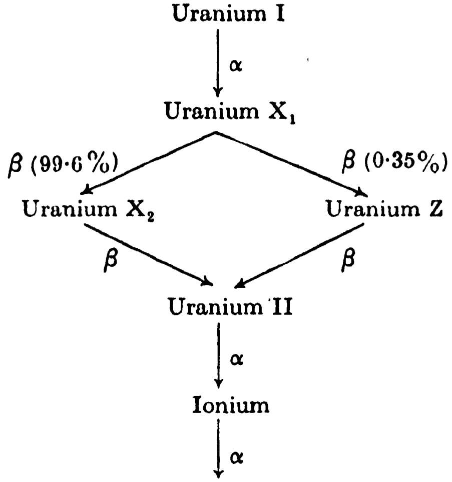
Uranium X_1 is an isotope of thorium (see Table 38, p. 152); it can therefore be most conveniently separated from solutions of uranium salts by precipitation with iron or zirconium hydroxide (see also p. 220).

	Uranium X_1	*Uranium X_2*	
Z	90 (= thorium)	91 (= protoactinium)	
A.W.	234	234	
T	24·5 days	1·14 minutes	
Radiation	β	β, γ	
μ	460	β	γ
		18	0·14
Parent element	U_I	UX_1	
Daughter element	UX_2	U_{II}	

Uranium X_2, an isotope of protoactinium, is obtained from solutions of uranium X_1 by electrolysis or by joint precipitation with tantalum oxide.

3. Uranium Y and Uranium Z

The genesis of these two products, both of which are found in uranium salts, is not yet perfectly clear. Uranium Y (T = 24·6 hours) is an isotope of thorium, and uranium Z (T = 6·7 hours) an isotope of protoactinium. Both are β-rayers (μ_{UY} = c. 54 and 600, μ_{UZ} = c. 100), and it follows from the smallness of their contribution to the total activity of uranium salts that they cannot occupy a place in the main disintegration series. Uranium Y is most probably a disintegration product of the uranium isotope ^{235}U and the parent of protoactinium, and hence of the actinium series (cf. Table 34, p. 143). On the other hand, uranium Z has hitherto been regarded as a daughter element of the dual β-disintegration of uranium X_1, but it is just possible that it may be a decay product of a uranium isotope other than ^{238}U (see p. 142). On the former view the beginning of the uranium-radium disintegration series would be represented schematically in the following way:



4. Ionium and Radium

Ionium, an isotope of thorium, has not yet been prepared in the pure state; a thorium preparation relatively rich in ionium (*re* its origin, see p. 151) was found to contain 30 per cent. of ionium, and yielded a combining weight of 231·51 (A.W. of ionium = 230, C.W. of thorium = 232·12).

	Ionium	*Radium*
Z	90 (= thorium)	88
A.W. . . .	230	226·05
T	$8{\cdot}3 \times 10^4$ years	1,590 years
Radiation . .	α	α (β) (γ)
R	3·2	3·39
Parent element .	U_{II}	Io
Daughter element .	Ra	Rn

We have already described the processes used in the preparation of radium. It is generally brought into the market in the form of the chloride or bromide, mixed with the corresponding salt of barium. One of the international radium standards is kept in Paris and consists of

22·23 mg. of anhydrous $RaCl_2$; the other is kept in the Vienna Radium Institute and contains 30·75 mg. of anhydrous $RaCl_2$. Both standards were prepared in 1934 by Hönigschmid, and are composed of the purest $RaCl_2$, free from every trace of barium. The price of radium preparations is determined by their content of the element radium. When the extraction of radium was limited to the small production at St. Joachimstal, the price of 1 mg. radium was between £20 and £25. Moreover, this price was not greatly altered in consequence of the more extensive production of the element in America. But with the discovery of the large mineral sources in the Belgian Congo and in Canada (Great Bear Lake), a marked diminution of the price has been effected. At the present time the price of radium is less than £5 per milligram.

Since radium, in spite of its rarity, has already been produced in quantities of several hundreds of grammes (see p. 211), it is no longer a difficult matter to determine the properties of this element by the methods usually used in chemistry and in physics. The evidence adduced from its spectrum and from the solubility of its salts, etc., all showed it to be a higher homologue of barium. Thus the solubility of its sulphate is smaller than that of any of the other sulphates of the alkaline earths, as is shown in Table 46.

TABLE 46

Solubility of the Sulphates of the Alkaline Earths

	Solubility at 25° C. in per cent. by weight (gm. of the sulphate in 100 gm. of the solution)
$CaSO_4$	$2{\cdot}1 \times 10^{-1}$
$SrSO_4$	$1{\cdot}5 \times 10^{-2}$
$BaSO_4$	$2{\cdot}3 \times 10^{-4}$
$RaSO_4$	$2{\cdot}1 \times 10^{-6}$

Table 47 gives the solubilities of the bromides, chlorides, and nitrates of barium and radium.

TABLE 47

Solubility of Barium and of Radium Salts (at 20° C., in per cent. by weight)

	Barium	*Radium*
Bromide . . .	51	41·4
Chloride . . .	26·3	19·7
Nitrate . . .	7·9	12·2

It is very surprising that the solubility of radium nitrate is greater than that of barium nitrate, and that in spite of this the radium becomes concentrated in the barium salt deposited when the mixed nitrates are subjected to fractional crystallization, just as in the case of the mixed bromides or chlorides.

Radium compounds decompose in the course of time under the action of their radiations; thus radium chloride gives off chlorine, and is partially converted into the chlorate and the oxide.

For the most part the salts of radium are available in quantities of only a few milligrams, and mixed with barium, so that it is most convenient to determine them quantitatively by the measurement of their radioactivity. In contrast to the method of weighing, the degree of purity of the radium salt is without significance in estimations by this method. We must compare the unknown preparation with a radium salt of known radium content which has been standardized directly or indirectly by means of the International Standards. When the intensity of the γ-rays is used as the basis of the measurement, the method has a further advantage in that the preparation can remain in a sealed-off glass tube, which is the usual mode of packing.† Very weak preparations must be brought into solution, and the most accurate method of estimating them is by the measurement of their radon content. In this method, when the radon is in equilibrium with the amount of radium present, it is driven into an electroscope that has been previously calibrated by known quantities of radon.

With the aid of this extremely sensitive method quantities of radium as small as 10^{-13} gm. can be measured accurately to within a few per cent., and hence it has been possible to establish the presence of traces of radium in all minerals. For further information see Chapter XXV (p. 274).

A weak β-radiation from radium is also mentioned in the table (p. 223). This does not originate in the nucleus, otherwise an isotope of actinium would be detectable as a disintegration product. It is of secondary origin, being released from the K- and L-levels by γ-rays (cf. p. 102). The value of the absorption coefficient is $\mu = 312$ cm.$^{-1}$ The γ-radiation proper to radium only amounts to approximately 1 per cent. of the total γ-radiation emitted by a radium preparation which is in equilibrium with its disintegration products. The α-activity of radium, exclusive of that of its disintegration products, is 0·56 times the α-activity of uranium, which of course always contains the two α-rayers U_I and U_{II} (in this connexion cf. p. 29).

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† It has been suggested that the unit of measurement for such γ-ray measurements should be called an 'eve'. This is the ionization produced per sec. in 1 c.c. of air at N.T.P. by the γ-rays from 1 gram of radium at a distance of 1 cm. (Cf. the unit of dosage on p. 57.)

5. Radon (Radium Emanation)

Radon is the heaviest of the rare gases, and because of its inability to enter into chemical reactions, and from the nature of its spectrum, it was possible at an early stage to allot it a corresponding place in the periodic classification. Later on it was obtained in sufficiently large quantities for the determination also of other of its constants. Its boiling-point has the value −62° C., its freezing-point −71° C., and its critical temperature +105° C. At room temperature, radon is distributed in the proportion of 1:3 between water and an equal volume of air in a closed vessel, so that its distribution coefficient α has the value 0·3. With increase in temperature the solubility decreases in strict accordance with Henry's law, as shown in Table 48. The solubility is also reduced by the addition of salts to the water; thus for uni-molar salt solutions at 18° C. the value of α is about 0·16. The distribution coefficient between blood and air amounts to 0·42 at body temperature. The human skin is somewhat permeable to radon, but the radon which enters the system during radium baths does so primarily by way of the lungs [3].

	Radon (Rn)
Z	86
A.W. . . .	222
T	3·825 days
Radiation . .	α
R	4·12
Parent element .	Ra
Daughter element .	RaA

TABLE 48

Distribution of Radon between Water and Air

Temperature in °C.	*Distribution coefficient (α) between water and air*
0	0·510
10	0·350
20	0·255
30	0·200
40	0·160
50	0·140
60	0·127
70	0·118
80	0·112
90	0·109
100	0·107

Organic liquids dissolve very much more radon than water; thus for acetone $\alpha = 6{\cdot}3$, petroleum $\alpha = 10$, hexane $\alpha = 17$, olive oil $\alpha = 28$, and so on. The coefficient of diffusion of radon in water at 14° C. amounts to 0·82 cm.^{2} day^{-1}. Also solid bodies like rubber, paraffin wax, platinum black, and especially charcoal adsorb radon very strongly. Under certain circumstances radium preparations occlude an appreciable fraction of the radon produced in them. Such salts which emanate badly can be improved by moistening, pulverizing, or by converting them into the hydroxide or halide. Solid radium salts emanate much more strongly when they are deposited on carrier substances with a large surface area. In this way it has been possible, by binding radium carbonate to iron hydroxide, to prepare sources of radon, the emanating power of which remained for years at over 90 per cent., a fact which is very welcome for various practical applications.

The volume of radon in equilibrium with 1 gm. of radium is 0·65 mm.^{3} ($= 6{\cdot}5 \times 10^{-6}$ gm.). This quantity of radon is called 1 'curie'; both it and the thousandth part of it ('milli-curie') are used as units of measurement for quantities of radon. For concentrations of radon the unit of measurement adopted is called 1 'mache'; thus spring-water has a radon concentration of 1 mache when the quantity of radon in 1 litre of this water would produce a saturation current of 1×10^{-3} electrostatic units, if it were transferred to an electroscope and if its radiation in all directions were utilized. One mache unit is equal to $3{\cdot}6 \times 10^{-10}$ curies per litre.† The radon content of strongly radioactive springs often amounts to several thousand mache units. Examples of this are contained in Table 49. Values for the radium content of sea- and river-waters are given on p. 275.

TABLE 49

Radon Content of Several Springs, expressed in Mache Units

Oberschlema	c. 3,000	Ischia (Old Roman Spring) . .	370
Joachimstal (Water in Mine) .	2,050	Gastein (Reissacher Stulm) . .	300
Brambach (Wettin Spring) .	2,000	Aix-les-Bains (Alum Spring) . .	56
Joachimstal (Spring Head) .	600	Carlsbad (Mühlbrunnen) . .	32

6. The Short-lived Active Deposit of Radium (RaA, RaB, RaC, RaC′, RaC″)

The first five disintegration products of the gas radon are isotopes of the metals polonium, lead, bismuth, or thallium, and since they are solids, they have the tendency to be deposited as soon as they are produced. Moreover, with the exception of a very small fraction they

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† In 1921 a new unit was suggested for the measurement of concentrations of radon: 1 'eman' = 1×10^{-10} curies per litre.

are positively charged, and hence they are deposited most easily on negatively charged surfaces. This fact is made use of in their preparation (cf. pp. 133 and 216). In consequence of their rapid transformation, most of our observations are made with mixtures of them, and we have already described the resulting complicated decay curves (p. 135 et seq.).

	RaA	RaB	RaC	RaC′	RaC″
Z . . .	84 (= polonium)	82 (= lead)	83 (= bismuth)	84 (= polonium)	81 (= thallium)
A.W. . .	218	214	214	214	210
T . . .	3·05 min.	26·8 min.	19·7 min.	10^{-6} sec.	1·32 min.
Radiation . .	α	β, γ	α, β, γ	α	β
R . . .	4·72		α 4·1	6·96	
μ . . .		β γ 890 230 77 40 13 0·57	β γ 50 0·23 13 0·127		
Parent element .	Rn	RaA	RaB	RaC	RaC
Daughter element	RaB	RaC	99·96% RaC′ 0·04% RaC″	RaD	RaD

Radium C also emits an exceedingly small number of α-particles (*c.* 22 per 10^6 particles of normal range 6·96 cm.) with ranges lying between the limits 8 cm. and 11 cm. (cf. Table 5, p. 27, and p. 100).

The absorption coefficient in lead of the hardest γ-rays from RaC, and hence also of a radium preparation that has not been freed from its disintegration products, has the value 0·533 cm.$^{-1}$

7. The Long-lived Active Deposit of Radium (RaD, RaE, RaF or Polonium)

The activity of a surface coated with the short-lived active deposit of radium does not diminish to zero, but shows a very weak residual radiation (β-radiation of RaD), to which new β- and α-rays are gradually added (RaE and RaF). Details on the curves of activity are given on p. 136. The whole of the activity does not disappear until after the disintegration of the RaF. The resulting substance is RaG, which emits no radiation because it is the stable end-product of the uranium-radium series, and according to the displacement laws it must be an isotope of lead. We have already mentioned in Chapter XV that it has been found in crystalline uranium minerals as 'lead of atomic weight 206' (see p. 150).

Radium D is also an isotope of lead, but owing to the presence of other types of lead in minerals, it cannot be obtained in the pure state from them. But it has been separated in quantities just visible and in a pure state from vessels in which about 1 curie of radon has been allowed to decay (cf. pp. 137 and 218); its electrochemical properties correspond exactly to those of ordinary lead.

Radium E is an isotope of bismuth. In view of its short period of life, it would appear to be impossible to obtain it in visible quantities. Absorption coefficients of 7·4 and 0·85 cm.$^{-1}$ have been found for the γ-radiation from radium E in copper. RaE has been produced artificially by bombarding bismuth with 5×10^6 e.V. deuterons [4].

	RaD	RaE	RaF
Z	82 (= lead)	83 (= bismuth)	84 (= polonium)
A.W. . . .	210	210	210
T	22 years	5·0 days	140 days
Radiation . . .	β, γ	β, γ	α
R			3·87
μ	β 5,500 γ 1·17	β 43 γ 0·24	
Parent element . .	RaC′	RaD	RaE
Daughter element .	RaE	RaF	RaG

Radium F, or polonium, is the most important representative of the element of atomic number 84; the other types of this element, the 'A'- and 'C′'-products, are still less stable. The γ-rays emitted by RaF are of very feeble intensity; 7 γ-quanta are emitted for every 10^6 α-particles. The average energy of these quanta amounts to 850×10^3 e.V. The chemical properties of this otherwise unknown element have to be studied with RaF, and in part are already very well known. It resembles very strongly its atomic analogues bismuth and tellurium, as is shown by the whole of its behaviour in analytical chemistry, by its electrochemical potential, which lies near to that of silver, and by its ability to form a gaseous compound with hydrogen. Like tellurium hydride, polonium hydride decomposes in the moist state in a few minutes. As already mentioned on p. 165, polonium occurs in the colloidal state in neutral or in weakly acid solutions.

B. The Actinium Series

1. Protoactinium and Actinium

	Protoactinium	*Actinium*
Z	91	89
T	$3{\cdot}2 \times 10^4$ years	*c.* 13 years
Radiation . .	α	β
R	3·67	
μ		unknown
Parent element .	UY (?)	Pa
Daughter element .	Ac	RdAc

It will be seen from the above table that uranium Y is presumed to be the parent of protoactinium; this is due to the fact that this arrangement fits in well with the displacement laws and with the quantitative relations known for the actinium series (see Chap. XIII), but it has not yet been possible to obtain direct proof of the genetic connexion between UY and Pa. The actinium series is present in a constant proportion in all uranium minerals, and is responsible for 4 per cent. of the total activity. In early geological times the proportion must have been much larger, in consequence of the relatively more rapid decay of actino-uranium (^{235}U), the parent of the actinium series. This parent element would then be a more prominent constituent in the mixed element uranium than it is to-day, when its relative abundance is estimated to be less than 1 per cent. and its rate of disintegration about 6 times that of uranium I.

The chemical properties of protoactinium are such as would be expected of a higher homologue of tantalum (see p. 212). The separation of protoactinium from tantalum is based on the fact that the basicity of Pa_2O_5 is stronger than that of Ta_2O_5, and is not much different from that of hafnium and zirconium. By making use of this analytico-chemical difference it has not only been possible to separate protoactinium from the accompanying tantalum, by precipitating the protoactinium along with zirconium phosphate, but also to prepare a few decigrams in the chemically pure state.

Actinium is a higher homologue of lanthanum, and since it is a new element it would be interesting if it could be prepared in the pure state. Hitherto this has not been possible, and owing to the short life of the element it is doubtful whether weighable quantities will be obtainable.

2. Radioactinium, Actinium X, and Actinium Emanation (Actinon)

	RdAc	AcX	AcEm (An)
Z	90 (= thorium)	88 (= radium)	86 (= emanation)
T	18·9 days	11·2 days	3·92 sec.
Radiation . .	α, β, γ	α	α
R	4·68 (α)	4·37	5·79
μ	β 175 γ 25 0·19		
Parent element .	Ac	RdAc	AcX
Daughter element .	AcX	An	AcA

Special attention is merited by one rather striking feature contained in the above table. AcX emits α-rays of smaller average velocity than RdAc, which has a longer period of life. This constitutes a breach of the general rule relating to the connexion between period of life and range (p. 146). As with the β-rays from Ra and RdTh, those emitted by RdAc may be assumed to be of secondary origin.

For the chemical separation of RdAc from Ac and simultaneously from AcX, we utilize the fact that thorium is readily precipitated in weakly acid solution by means of thiosulphate or hydrogen peroxide. In order to obtain a visible deposit we may add a trace of a thorium salt, or, still better, of a salt of the non-isotopic element zirconium (cf. p. 219 et seq.) before the precipitation. In this way we obtain AcX together with Ac in the filtrate, and it may be freed from Ac by repeated precipitation with ammonia, remembering of course that we must add a little iron or similar element before each precipitation with ammonia, in order to obtain a deposit the anions (OH′-groups) of which then act as carriers for the Ac (cf. the rule on p. 220). We may obtain AcX in a very pure form by means of recoil from solid preparations of RdAc (cf. p. 218).

Like radon, actinon is an inert rare gas, the physical constants of which (coefficient of diffusion, solubility, etc.) have not been determined with any great accuracy, owing to its extremely short period of life and the correspondingly small quantities of it available. In view of the isotopy of the two emanations, however, we may assume with confidence that the properties of An are identical with those of Rn.

3. **The Active Deposit of Actinium** (AcA, AcB, AcC, AcC′, AcC″)

	AcA	AcB	AcC	AcC′	AcC″
Z . . .	84 (= polonium)	82 (= lead)	83 (= bismuth)	84 (= polonium)	81 (= thallium)
T . . .	2×10^{-3} sec.	36·0 min.	2·16 min.	5×10^{-3} sec.	4·76 min.
Radiation . .	α	β, γ	α, β	α	β, γ
R . . .	6·58		5·5 (α)	6·6	
μ . . .		β large γ 120 31 0·45			β 44·5 γ 0·198
Parent element .	An	AcA	AcB	AcC	AcC
Daughter element	AcB	AcC	0·16% AcC′ 99·84% AcC″	AcD (?)	AcD

A comparison with the table on p. 228 reveals immediately the great similarity, as regards their sequence and radiation, between the individual members of the active deposit of actinium and the corresponding members of the short-lived active deposit of radium. Apart from the different numerical values of T, R, and μ, the following important differences exist. In the first place, with AcC the majority of the atoms disintegrate with emission of α-radiation into AcC″, whereas in the case of RaC practically all the atoms are transformed into RaC′ with emission of β-rays; in the second, AcD is already the stable end-product of the actinium series, whereas RaD, although it is much more stable than the previous members of the series, still possesses the character of a radio-element, and is the first member of the 'long-lived active deposit' of radium, which has no analogue in the actinium series.

C. The Thorium Series

1. Thorium, Mesothorium 1, and Mesothorium 2

	Th	$MsTh_1$	$MsTh_2$
Z	90	88 (= radium)	89 (= actinium)
A.W. (and C.W.) .	232·12	228	228
T	$1{\cdot}34 \times 10^{10}$ years [5]	6·7 years	6·13 hours
Radiation . . .	α	(β)	β, γ
R	2·59 [6]		
μ			β: 40–20 γ: 26 0·116
Parent element . .		Th	$MsTh_1$
Daughter element .	$MsTh_1$	$MsTh_2$	RdTh

The element thorium is the parent of a radioactive series which is completely independent of the uranium-radium and the actinium disintegration series. This is evidenced by the fact that the contributions of the two series towards the activity of various minerals fluctuate between very wide limits. Thus we are familiar with uranium minerals that are practically free from thorium and its decay products, and, conversely, there are thorium minerals which contain very little uranium, and hence also very small quantities of ionium and radium. The chief source of thorium is monazite sand, which contains up to 18 per cent. ThO_2. A few rare minerals are much richer in thorium, such as the thorianite found in Ceylon, which may contain over 50 per cent. ThO_2. A résumé of the composition and properties of the most important thorium minerals is given in Table 52 on p. 273. The separation of thorium from the other rare earths is not a simple matter, but information on the preparation of pure thorium is contained in chemical text-books. In radioactive investigations thorium is generally used in the form of nitrate or hydroxide. The nitrate is used when we require a soluble salt for the purpose of separating out ThX, and the hydroxide for the collection of thoron, owing to the fact that it 'emanates' freely.

We have already spoken (Chap. XXII, p. 212) about the preparation of *mesothorium*, and of the impossibility of obtaining it completely free from radium. We can determine the approximate radium content of mesothorium preparations on the market without opening them, by carrying out absorption measurements of the γ-rays for various thicknesses of the absorbing metal sheets (see p. 55). More reliable values are obtained by determining the amount of radon produced in them.

We are unable to detect the rays emitted by mesothorium 1, but from the fact that the succeeding product mesothorium 2 has an atomic number 89, we must conclude that each atom of the former loses a β-particle when it is transformed into an atom of the latter. The activity is due exclusively to the radiation from the disintegration products, especially that of mesothorium 2, which, owing to its short half-value period, is present practically in its equilibrium amount even after a period of 3 days. An appreciable quantity of radiothorium will be present in older preparations of mesothorium, since the half-value period of RdTh is only 1·9 years. In consequence of this we have to take account of the likewise very penetrating γ-radiation from ThC″. Fig. 48 indicates the manner in which the resulting total activity of a 'mesothorium preparation' alters in the course of years.[†]

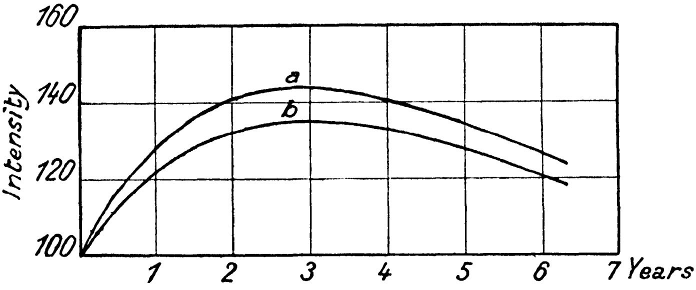

FIG. 48. Variation of the γ-Activity of Mesothorium Preparations with Time.

Curve *a* was obtained with freshly prepared mesothorium free from radium, whilst curve *b* was obtained with a new preparation of mesothorium containing radium (cf. footnote on p. 55). In these measurements the rays were passed through lead filters of thickness 3·3 mm. In both cases the maximum intensity of the radiation was attained after a period of about 3 years, but the increase was greater for the preparation free from radium than for that containing radium. In the former case the activity rose from 100 to 143·8, or by about 44 per cent., whereas in the latter case it rose from 100 to 135, or by 35 per cent. If the measurements had been carried out with lead filters 5 mm. instead of 3·3 mm. thick, the form of the curves would have been essentially the same, but the rise would have been slightly more pronounced for the mesothorium containing radium, owing to the fact that the γ-radiation from radium relative to that from mesothorium is somewhat less effective after passing through 5 mm. of lead than after passing through 3 mm.

The amount of MsTh available by weight is always extremely small,

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[†] Since the hardnesses of the γ-radiations from $MsTh_2$ and ThC″ are different, the measured proportions of the two radiations also depend somewhat on the choice of the apparatus used for the measurements (thickness of the absorbing layers).

for $2{\cdot}7 \times 10^9$ gm. thorium are required to produce an equilibrium amount of 1 gm. MsTh_1. On the other hand, the γ-activity of 1 gm. radium is equivalent to that of about $\frac{1}{500}$ gm. mesothorium, and hence to the quantity in equilibrium with about 5×10^6 gm. thorium.

2. Radiothorium, Thorium X, and Thorium Emanation (Thoron)

	Radiothorium	*Thorium X*	*Thoron*
Z	90 (= thorium)	88 (= radium)	86 (= radon)
A.W. . . .	228	224	220
T	1·90 years	3·64 days	54·5 sec.
Radiation . . .	α, β	α	α
R	4·02 (α)	4·35	5·06
μ	β 420		
Parent element . .	MsTh_2	RdTh	ThX
Daughter element .	ThX	Tn	ThA

The β-rays from RdTh do not originate in the nucleus, but are of secondary origin (cf. p. 102), like those from Ra and RdAc. They are probably released from the *L*- and *M*-levels by a γ-radiation which has not yet been detected directly.

We have already mentioned the method of preparing radiothorium in Chapter XXII. ThX may be obtained periodically from a preparation of RdTh by precipitating this together with iron, aluminium, or similar elements by means of ammonia, when the ThX remains in the filtrate (cf. the method described above for the preparation of the isotope AcX). Radiothorium and thorium X are chiefly used as sources of thoron and of its short-lived disintegration products. It is advisable to maintain preparations that have been made for this purpose permanently in a moist condition, because then they allow the emanation to escape much more readily.

3. The Active Deposit of Thorium (ThA, ThB, ThC, ThC′, ThC″)

	ThA	ThB	ThC	ThC′	ThC″
Z . . .	84 (= polonium)	82 (= lead)	83 (= bismuth)	84 (= polonium)	81 (= thallium)
A.W. . .	216	212	212	212	208
T . . .	0·14 sec.	10·6 hours	60·5 min.	c. 10^{-11} sec.	3·1 min.
Radiation . .	α	β, γ	α, β	α	β, γ
R . . .	5·68		4·78 (α)	8·62	
μ . . .		β 153 γ 160 32 0·36	β 14·4		β 21·6 γ 0·096
Parent element .	Tn	ThA	ThB	ThC	ThC
Daughter element	ThB	ThC	65% ThC′ 35% ThC″	ThD	ThD

Comparisons of the accompanying table with that for the active deposit of actinium and with that for the short-lived active deposit of radium (pp. 231 and 228) reveal the great similarity between these parts of the three disintegration series. Characteristic of the thorium series is the branching ratio of the C-product. In this case comparable amounts disintegrate with the emission of α- and of β-rays, whereas with AcC almost the whole follows the α-branch, and with RaC the main disintegration is attended by the emission of β-rays. According to the displacement laws it is to be expected that the two branch series will lead to the same end-product, the lead-isotope ThD, and as a matter of fact a type of lead with the atomic weight calculated for ThD has been found in thorium minerals (cf. p. 151). It has not yet been established whether both types of lead, produced in the proportion by weight of 35:65, are stable, or whether one of them is transformed into an isotope of bismuth with emission of β-rays (cf. p. 144).

Although the A-product is of longer life than in the case of the actinium series, it has likewise not yet been possible to separate it from the emanation. Both products reveal themselves in the fact that the scintillations produced by the emanations are always observed to occur in pairs. In the case of actinium ($T_{\mathrm{AcA}} = 2 \times 10^{-3}$ sec.) the two members of such a pair of scintillations appear simultaneously, whereas in the case of thorium ($T_{\mathrm{ThA}} = 0{\cdot}14$ sec.) a distinct interval between the two scintillations is recognized, albeit a short one.

ThB and ThC are particularly suitable for use as indicators in physical and in chemical investigations, and have often been used for this purpose (cf. Chap. XVIII). This is due to the fact that they are relatively stable and their half-value periods are so different that they can conveniently be determined quantitatively, one along with the other.

The active deposit of thorium also emits α-particles of ranges 9·8 and 11·6 cm. For every 10^6 α-particles of range 8·62 cm. (ThC′) there are respectively 65 and 180 of these particles of long range (cf. p. 27).

D. POTASSIUM, RUBIDIUM, AND SAMARIUM

	Potassium		*Rubidium*	*Samarium*
Z	19		37	62
C.W.	39·096		85·48	150·43
A.W.	39; *40*†; 41		85; *87*†	144; 147; 148; 149; 150; 152; 154
T	$1{\cdot}9 \times 10^9$ years		5×10^{10} years	$1{\cdot}02 \times 10^{12}$ years
Radiation	β	γ	β	α
μ	75; 29	0·59 in lead	700; 140	
R				1·15 cm.

† Radioactive isotope. The half-value periods given for potassium and rubidium are

From amongst the chemical elements that have been known for a long time it has been found by very careful measurements that there are three, apart from uranium and thorium, which emit a constant but very weak radiation. These are the elements potassium, rubidium, and samarium. There is some evidence also that neodymium emits β-rays.

Potassium and rubidium are β-rayers, and although the rays emitted by potassium are the more penetrating, the number of β-particles ejected from equal quantities by weight is markedly greater in the case of rubidium. Measurements with a tube counter have yielded for the ratios of the numbers of β-particles emitted in the same time by equal amounts of potassium, rubidium, and uranium the values 1:16:500. In this estimate account is taken of only one of the β-raying components resulting from uranium, viz. UX_1. The limit of hardness of the softer component of the β-radiation amounts to 400×10^3 e.V. for potassium and to 100×10^3 e.V. for rubidium, whereas the limit of hardness of the fast β-rays from potassium is situated at 700×10^3 e.V. and from rubidium at 250×10^3 e.V. The γ-radiation from potassium is almost as hard as the hardest γ-radiation emitted by radium (RaC), but its intensity is extremely small. The ratio between the intensities of the γ-radiations from 1 gm. potassium and from RaC (in equilibrium with 1 gm. radium) has the value $1:3 \times 10^{10}$. In the disintegration of 100 atoms of potassium only three γ-quanta are emitted, each with an energy of 2×10^6 e.V.

By means of experiments devised for the purpose it has been shown that the radioactivity of potassium and rubidium compounds is strictly proportional to their respective potassium and rubidium contents, and that the radiation has the same intensity at the temperature of liquid air as at room temperature. There can be no doubt, therefore, that we are here dealing with the phenomenon of radioactive disintegration. It is a striking fact that, apart from samarium (see later), the elements potassium and rubidium stand in an isolated position, for all the other natural radio-elements belong to disintegration series consisting of many members. The discovery of artificial radioactivity, however, has thrown important light on this remarkable phenomenon (cf. pp. 115 and 238).

In order to decide which of the isotopes of potassium is responsible for the emission of β-rays, a partial separation of the potassium isotopes was undertaken, and both the combining weight and the activity of the 'heavier' fraction obtained were compared with those of ordinary potassium. The determination of the combining weight of the heavy

those of ^{40}K and ^{87}Rb; in the case of samarium the radioactive isotope is not known, and hence the period given in the table is that which samarium would have if the whole of the mixed element were radioactive.

fraction revealed an increase in the concentration of ^{41}K of about 7 per cent., whereas the β-activity of the heavy fraction was found to have increased by only about 4 per cent. as compared with ordinary potassium. Now for the same conditions of experiment the gain in the concentration of a potassium isotope can depend only on the difference in the mass of that isotope from that of the isotope ^{39}K. From the fact that it is more difficult to increase the concentration of the radioactive isotope than it is to increase that of ^{41}K, it follows that the mass of the radioactive isotope must differ by less than 2 units from that of ^{39}K, so that it must have the value 40. This conclusion is supported by the fact that it has not been possible, either by the mass-spectrograph or on the basis of atomic-weight determinations, to detect the presence of a calcium isotope of mass 41 in potassium minerals of great geological age. It is interesting to note that the presence of ^{40}K in ordinary potassium has been established to the extent of about 1:8,000 by means of a mass spectrograph of high resolving power [7], and that by use of an instrument of high intensity it has also been possible to separate in minute amounts the three isotopes of potassium. Measurements of their activity show not only that ^{40}K is radioactive, but that it is the isotope responsible for the two known groups of β-rays emitted by potassium [8]. From the above-mentioned concentration of ^{40}K in the mixed element potassium, and the apparent half-value period of potassium, $1{\cdot}5 \times 10^{13}$ years, which follows from the number of β-particles emitted annually by 1 gram of potassium, the half-value period of ^{40}K works out to be *c.* 2×10^9 years.

By analogy with potassium it seemed not unlikely that the radioactivity of rubidium would be due to an isotope of mass 86, present in quantities too small to be detected by the mass spectrograph, but this view is not substantiated by recent experimental work. Investigations carried out with a lithium mica from Manitoba, Canada, showed that it contained 2–3 per cent. of rubidium and a slight amount of strontium. The strontium was carefully extracted from about 1 kg. of this mineral, and about 250 mg. of strontium carbonate were obtained; calculation indicated that at least the major part of this strontium must have been produced in the mineral by the disintegration of rubidium. A careful mass-spectrographic examination of the strontium preparation has shown that it consists almost wholly (99·7 per cent.) of the strontium isotope ^{87}Sr,† and the conclusion seems inevitable that the isotope of rubidium responsible for the radioactivity of this element is ^{87}Rb [9]. From this work it also appears that the value of the apparent half-

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† Ordinary strontium has the following approximate composition, as determined by the mass spectrograph: 10 per cent. of ^{86}Sr, 7 per cent. of ^{87}Sr, and 83 per cent. of ^{88}Sr (see p. 183).

value period of rubidium previously accepted ($4 \cdot 3 \times 10^{11}$ years) is too high, and that the true value lies nearer 2×10^{11} years. In view of the abundance of ^{87}Rb in ordinary rubidium (27 per cent.), the value of the half-value period of ^{87}Rb must be about 5×10^{10} years.

Until comparatively recently it was quite impossible to explain the existence outside the radioactive disintegration series of elements which emit β-rays spontaneously [10]. But that difficulty has been removed since the discovery of artificial radioactivity. Substances that have been rendered radioactive by artificial means mostly emit β-rays, and usually have a relatively short period of life. For this reason isotopes that were rendered active in the distant past by the action of neutrons and other suitable agencies in the cosmos are no longer to be found to-day in terrestrial materials or in meteorites, even by the use of the mass-spectrograph. But in the case of potassium and rubidium, thanks to the longer periods of life of ^{40}K and of ^{87}Rb, we are at the present time able to detect measurable quantities of these isotopes.

As a result of the transformation of ^{40}K the chief isotope of calcium ^{40}Ca will be produced, just as the disintegration of ^{87}Rb results in the production of the isotope ^{87}Sr of strontium. This will be clear if we recall that the emission of a β-particle increases the nuclear charge by unity, so that from potassium ($Z = 19$) the element calcium ($Z = 20$) is produced, and from rubidium ($Z = 37$) the element strontium ($Z = 38$). On the basis of these results, it was to be expected that calcium would accumulate in potassium minerals and strontium in rubidium minerals in the course of geological time. It is worthy of note that, in so far as evidence is available on this point, it lends support to the conclusion that ^{40}Ca, at least in part (cf. p. 124), is the product resulting from the disintegration of potassium, and we have already seen that ^{87}Sr is the product of the disintegration of rubidium.

Since the β-radiation of rubidium is distinctly softer than that of potassium (see previous table), it would appear that an eventual β-radiation from caesium might have escaped detection, in consequence of its still greater softness and feeble intensity. It can be asserted with confidence, however, that the total ionization which might be produced by caesium (inclusive of eventual α-rays) has a value less than $\frac{1}{40}$ that of rubidium, the corresponding total ionization in the case of sodium being certainly less than $\frac{1}{50}$ that from potassium.

Samarium is the only element outside the radioactive disintegration series that emits α-rays [11]. These have a range of 1·15 cm. at 15° C. and 760 mm. Hg. One gram of samarium emits 88 ± 5 α-particles per second, and is accordingly 270 times less active than one gram of uranium. The half-value period of samarium ($1 \cdot 02 \times 10^{12}$ years) given in the table has been calculated on the assumption that, in the course of time, all the atoms of samarium take part in the disintegration process. Presumably only one of the 7 known isotopes of samarium emits α-particles, but it is not yet known which one is responsible. In order to obtain the true half-value period of the radioactive type of samarium, it will be necessary to multiply the above half-value period by the concentration of the active component in the mixed element samarium.

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XXIV

EFFECTS OF THE RAYS FROM RADIUM

THE effects produced by the rays from radium may be grouped in the following way: 1. Photographic Action; 2. Ionization; 3. Excitation of Luminescence; 4. Development of Heat; 5. Chemical Effects; 6. Colloido-chemical Effects; 7. Biological and Therapeutic Effects; 8. Mechanical Effects; 9. Atomic Disruption.

1. Photographic Action

We mentioned in the Introduction (p. 1) that the rays emitted by radioactive substances exert a similar action on a photographic plate to that produced by ordinary light; after development, the plate becomes blackened where the rays have been incident. When radioactive research was in its infancy, and before the methods of measurement based on the ionization of air had been developed, the photographic plate was the main instrument used in the study of the new phenomena, and even to-day it is by far the most suitable piece of apparatus for certain experiments. Thus, in experiments on X-ray spectra it has been found that photography enables us to work in a much simpler and more exact manner than with ionization chambers, which were originally used. Likewise, the 'spectra' of β-rays, obtained by deflexion in a magnetic field, are best registered on a photographic plate (cf. Fig. 17, Plate I). It can also serve the purpose of enabling us accurately to follow the tracks of α-particles. If we place the point of an activated needle in contact with a photographic plate and allow it to act for a short time, we find on development a very tiny blackened patch on the plate, which appears under the microscope as a number of resolved radial tracks, consisting of a series of silver grains. They are caused by the α-rays which have traversed the outermost film of the plate at grazing incidence. The rectilinear tracks of α-rays can also be detected in this way, as well as with the help of cloud-chamber photographs (cf. pp. 19 and 32). Under favourable circumstances as many as 20 silver grains may be rendered capable of development by an α-particle of 7 cm. range, so that the number of impacts with silver particles in the plate is 20. This follows from the fact that it has been possible to show that every grain struck by an α-particle is capable of development, whereas on the average 6 to 8 β-particles are necessary to make a silver halide nucleus capable of development. The blackening caused by α-rays is proportional to the product of the intensity and the exposure time, so that Bunsen and Roscoe's relation can be applied to determine the intensity

PLATE V

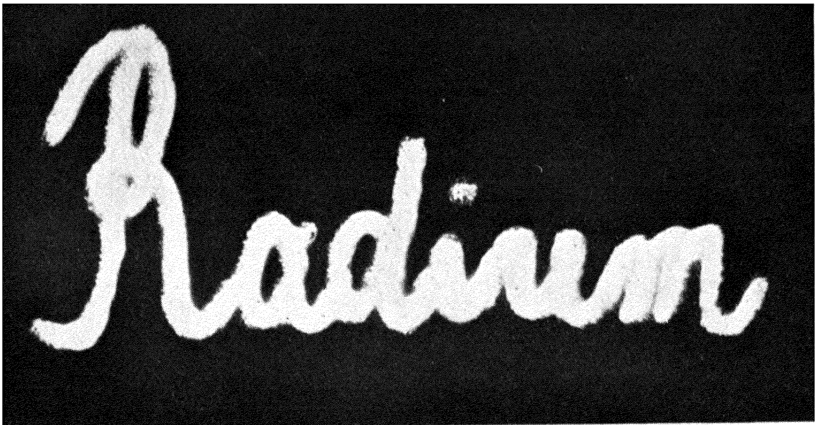
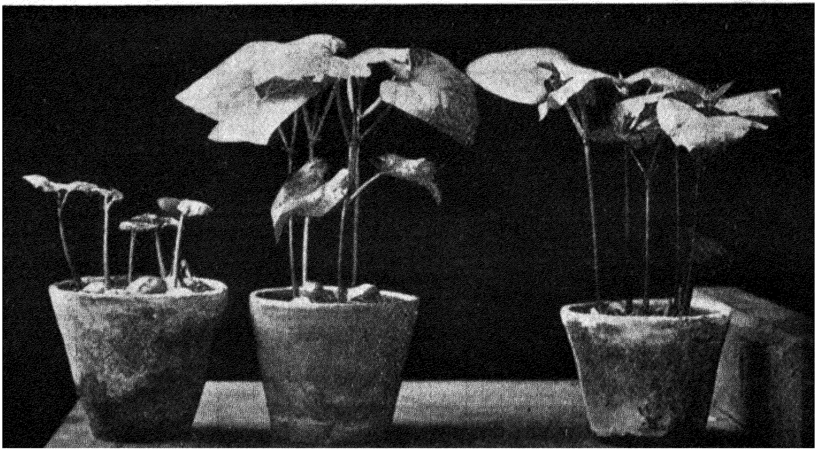


Fig 49 (p. 241). Writing on a Photographic Plate by means of Radium.



(*a*) (*b*) (*c*)

Fig. 51 (p. 259). Action of Radon on Shoots of *Phaseolus multiflorus*. [(*a*) Effect of a large quantity of radon; (*b*) Effect of a small quantity of radon; (*c*) Control specimen, without radon.]

of the radiation from the degree of blackening produced. In mineralogy it is often useful to be able to establish the position of a radioactive inclusion in a mineral; a photographic plate laid on a thin section of the mineral will reveal even the smallest active inclusion, provided the experiment be carried out over a sufficient length of time. In the chapter on 'Radioactive Indicators' we have also mentioned an example (p. 175) in which the photographic plate is able to achieve more than the ionization chamber. Nevertheless, the sensitiveness of the photographic method, say for the weak β-rays from radium, is distinctly smaller than that of electroscopic measurements.

Since β- and γ-rays are able to pass through solid substances, in a manner dependent on the density of these substances (see pp. 39 and 51), the possibility suggests itself of producing radiograms by means of radium rays, similar to those produced by X-rays. They are, however, much less suitable for this purpose. In the case of the β-rays the reason is to be found in the fact that, in consequence of marked scattering effects, the edges of the objects radiographed always appear blurred; but even if we deflect the β-rays by means of strong magnetic fields, so that only the γ-rays strike the photographic plate, the radiograms obtained will still be worse from the photographic viewpoint than those obtained by X-rays, because, owing to the greater penetrating power of the γ-rays, the contrasts between the radiographed objects of greater and lesser density are smaller. γ-radiograms are of service, however, in the examination for flaws of large metallic objects, too thick for the application of X-rays.

The length of exposure necessarily varies within wide limits, depending on the strength of the radioactive substance. Fig. 49 (Plate V) shows writing which was obtained by using a preparation of 40 mg. of radium enclosed in a glass tube. This served as a pencil, and was drawn slowly to and fro over the photographic plate, which was wrapped in black paper. The β-rays from the preparation were almost exclusively responsible for the effects in this experiment.

At those places on the gelatine film that are most strongly bombarded by α-particles indentations occur; the swelling capacity of the gelatine is reduced. Moreover, marked solarization effects are observed on those parts of the plate that have been subjected to the action of strong sources of α-rays. When the duration and intensity of the action of the rays on the photographic plate exceed certain values, it is found that the plate is blackened even before it is developed.

2. Ionization

This action of the rays from radium has already been described (p. 7 et seq.), and we have seen that the ionization of the air forms the basis of the whole of the technique of radioactive measurements. It may be added, however, that solid and liquid dielectrics also become conducting under the influence of the rays from radium, in a similar manner to gases. The behaviour of hexane, a liquid hydrocarbon with the formula C_6H_{14}, is of interest; the ions produced in this liquid show the usual mobility of electrolytic ions, but since the number of ions produced in unit time is only small, the phenomenon of the saturation current manifests itself here also, just as in the ionization of gases (cf. the first footnote on p. 10). We may assume that the small conductivity always shown by hexane is to be attributed wholly or in part to the action of the natural penetrating radiation (see p. 278).

It has also been shown that the conductivity of pure water increases under the action of the radiation. But in this case it is not quite certain whether we have here an actual increase in the conductivity of the water itself, or whether the rise is due to the solution of some of the material of the walls of the vessel under the action of the rays, and the consequent addition of a trace of an electrolyte. In view of the extreme sensitiveness of water towards small amounts of impurity, the latter possibility cannot very readily be excluded.

The ionizing action of the rays from radium is sometimes also used in other physical measurements apart from radiological ones. By this means, for instance, we can investigate the contact potential between metals. Moreover, it is of especial use in measurements in atmospheric electricity, where we can replace flame electrodes by ionizing discs activated with, say, polonium, in order to measure the potential of the surrounding air.

3. Excitation of Luminescence

The rays from radium, themselves invisible, are able to excite substances to emit visible light, as already briefly mentioned on p. 1. This is most strikingly manifested in the case of the phenomenon of *autoluminescence.* Thus, when we subject a barium salt containing radium to fractional crystallization, the mixed barium and radium salt becomes self-luminous in the dark when the concentration of radium in the mixture has reached a certain value. The colour depends on the relative proportion of radium to barium present in the mixture, and can, in fact, serve for the approximate estimation of the radium content of the preparation. When a 100 per cent. preparation of radium chloride is fused and thus completely freed from water, it emits a bluish light which is so intense that it can be seen even in bright daylight. Investigations of the spectrum of this light have shown that it consists of the band spectrum of nitrogen. Correspondingly, the helium lines are obtained in an atmosphere of helium. The phenomenon of self-luminescence (i.e. really the excitation of fluorescence in the neighbouring gas) has also been observed with strong solutions of radium salts, and with strong preparations of polonium, ionium, mesothorium, and actinium.

With weak preparations we can obtain the phenomena of luminescence only by bringing them near to substances which possess marked fluorescent properties. As is well known, barium platino-cyanide ($BaPtCy_4+4H_2O$), zinc blende, and zinc silicate are very suitable for this purpose. The first is used particularly for the detection of X-rays, and likewise for γ-rays, whilst the last two show the phenomenon of 'scintillation', and enable us to detect the incidence of individual α-particles. Each α-particle gives rise to a single flash of light when it strikes a screen coated with powdered zinc blende, and by suitably choosing the strength of the α-ray preparation, we may observe, particularly with a lens, a highly characteristic phenomenon—the luminous field appears to be in a state of agitated motion, as a result of the ever-changing incidence of individual luminous points. This apparatus can be made very simply, and is known as a 'spinthariscope'. The duration of an individual flash of light is only 10^{-4} seconds.

Special precautions are necessary for the preparation of samples of zinc blende which fluoresce satisfactorily. The amorphous ZnS powder obtained by precipitation from an aqueous solution does not show phosphorescence, but it can be transformed into the crystalline form by heating to redness. Before the precipitation we must either ensure the presence of an excess of the zinc salt, or we must add a foreign heavy metal like copper, because even the crystalline blende is capable of phosphorescence only when 'nuclei' are available (see below). The far-reaching conclusions resulting from the fundamental investigations on the scattering of α-particles and on the disruption of atoms were based initially on the appearance of a few flashes of light on zinc sulphide screens, in unexpected directions and at unexpected distances. This emphasizes the great importance which attaches to the preparation of good zinc sulphide screens for scientific work.

Preparations of zinc sulphide with which a certain quantity of a radioactive substance has been mixed are of practical importance, for they exhibit a more or less permanent luminescence, which is independent of external sources of energy. These radioactive 'luminous substances' differ from the phosphorescent substances formerly used, in that they do not require previous illumination before they luminesce. They are used for coating the pointers and figures of clocks, watches, and compasses, and for rendering visible such things as signs (e.g. in theatres), bait on fishing-hooks, and so on. The most rational α-rayer to use for this purpose is radiothorium, since the luminescing power of every zinc sulphide preparation appreciably diminishes even after the lapse of a few months. For this reason there is no special point in using long-lived substances like radium or mesothorium, although radium is at present less expensive than radiothorium. In practice, one gram of zinc sulphide is mixed with a quantity of active material, the intensity of the γ-radiation from which is equivalent to 10^{-4} to 10^{-1} mg. of radium. When a good sample of zinc sulphide is available, several per cent. of the energy of the α-rays is transformed into light, and this is fortunately distributed for the most part over the visible spectrum, with a maximum at $550\,\mathrm{m}\mu$ (see p. 30).

The above-mentioned diminution with time of the luminescing power of preparations of zinc sulphide is most probably to be explained on the assumption that the luminescence originates in the 'centres' or nuclei that are postulated in all 'phosphors'. In zinc sulphide these centres are present presumably only in relatively small numbers, about one in every thousand ordinary molecules, and they are gradually destroyed by the action of the radiations from the radioactive substance.

Apart from barium platino-cyanide, zinc blende, and zinc silicate, a large number of substances fluoresce more or less strongly under the action of the rays from radium; thus the mineral willemite (Zn_2SiO_4) emits green light, kunzite (a silicate of lithium and aluminium) emits red light†, scheelite ($CaWO_4$) fluoresces brilliantly and diamond much less strongly, both emitting blue light. With these last two substances scintillations can also be observed when α-particles are incident upon them. Moreover, most benzene derivatives luminesce under the action of the rays from radium, particularly those with many rings and the derivatives of salicylic acid, e.g. salipyrin (= acid salicyl-antipyrin). Furthermore, the crystalline lens, the vitreous fluid, and the retina of the eye fluoresce under the action of γ-rays; this is the reason why even blind persons, provided the retina and optic nerve are not damaged, are able to experience the sensation of light when a radium preparation is brought near their eyes. In such experiments great care should be exercised, owing to the possibility of physiological damage being done to the tissues by the rays from radium (see § 7).

Most fluorescing substances become coloured under lengthy exposure to the rays, but simultaneously the fluorescence diminishes and 'fatigue' occurs. This fact is both of practical and of theoretical interest. By heating, often only slightly above room temperature, or by illumination with light, the colouring can frequently be annulled, and at the same time the ability to fluoresce restored. This decoloration by means

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† Kunzite, and in a lesser degree also fluorspar and a few other crystals, can be excited to show luminescence very strongly when they are first rayed by γ-rays and later with ordinary light.

of heat or light is generally accompanied by luminescence and the emission of electrons (thermoluminescence and photo-electric effect), as, for instance, with kunzite, the original rose colour of which is changed to green by the rays, or with fluorspar, which is transformed from a green to a blue colour by the action of the rays. New vessels and tubes of glass luminesce much more strongly under the action of radioactive solutions or gases than those that have been in use for a long time; this is a striking illustration of the fact that glass fatigues or loses its ability to fluoresce when subjected to the action of the rays. Glass apparatus that has been exposed to the rays for a long time always has a brown or violet colour (see p. 248), and also shows very clearly the phenomenon of thermoluminescence (with the well-known characteristic green glow of X-ray tubes). Quartz that has assumed a violet colour also shows the same luminescence during its decoloration by means of heat.†

The close connexion between the phenomena of luminescence and colour changes can be readily understood from the theoretical point of view. Detailed investigations in this field have been undertaken with various salts, but primarily with natural rock salt, and they have led essentially to the following result. When NaCl is coloured by 'illumination' with β- or γ-rays, the primary process consists in the absorption of a quantum of radiation by a chlorine ion, whereby the superfluous electron responsible for the negative charge of the chlorine ion is removed and transported to one of the neighbouring positive Na-ions. The latter is neutralized in this way and transformed into an atom of sodium, which has the ability to absorb visible light. The resulting colour will depend on whether and how strongly the discharged sodium ions (i.e. atoms) are influenced by lattice forces; if they are freely mobile and unite to form larger complexes, a process which is favoured by a moderate rise of temperature, they then absorb yellow light in the neighbourhood of the D-line, as we should expect from theory, and the rayed rock-salt assumes a blue-violet colour.‡ By means of experiments on the influence of pressure applied to one side of the crystal it has been possible to show that the process above described takes place especially at disturbed places in the crystal lattice. For the same reason, alkali chlorides that have been crystallized from a fused mass are more susceptible to colour changes than are the more regularly crystallized

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† Thermoluminescence and 'phosphorescence' cannot be sharply differentiated; thus doubly refracting calcspar that has been coloured yellow phosphoresces with emission of red light even at ordinary temperatures.

‡ The often noted agreement in the colour of rayed salts with that of their colloidal liquid solutions (sols) is also based on a separation of the metal of this nature; thus, under the influence of the rays from radium, borax beads to which a little lithium has been added become brown, with sodium they become violet, with potassium blue, and with rubidium greenish blue, and these are just the colours of the corresponding sols.

salts obtained by crystallization from solutions. When the raying process is interrupted and the coloured salt is heated, the valency electron of the sodium atom returns to the chlorine atom, whereby the ionic lattice is reformed and decoloration takes place. This return of the electron can give rise to the emission of light in different ways, and on these lines it is possible to explain the thermoluminescence produced by heating coloured salts and glasses. The phenomena of the emission of electrons, decoloration by the action of light, and in particular also the feeble fluorescence of strongly coloured glasses all find a satisfactory explanation within the framework of this theory, but we shall not discuss them in further detail here.

4. Development of Heat

The following details will serve to amplify the information already given on p. 1.

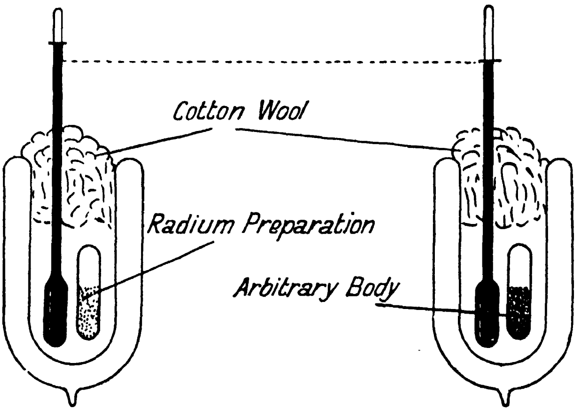

FIG. 50. Heat Development of a Radium Preparation.

A simple experimental arrangement that enables us to demonstrate the continuous liberation of heat by radium preparations is represented diagrammatically in Fig. 50. A radium salt contained in a glass tube is situated in the left of two identical Dewar vessels, each provided with a thermometer, and in the right-hand vessel is a similar tube containing the same quantity of a barium salt. The temperature in the left-hand vessel is always found to be somewhat higher than in the right-hand vessel. If the salt introduced contains half a gram of the element radium, the difference in temperature may amount to as much as 5° C., and is only limited by the magnitude of the unavoidable loss of heat to the exterior. Correspondingly, the mercury thread in a Bunsen ice calorimeter, by means of which the amount of ice melted per unit of time is measured, moves constantly with uniform velocity.

The amount of heat is generally determined quantitatively by evaluating the amount of electrical energy which produces the same quantity of heat as the preparation concerned (method of compensation). In setting up the experiment, of course, care must be taken to ensure that all the rays emitted are absorbed within the calorimeter, with the exception of a fraction of the γ-rays, for which a correction can be applied. In this manner it has been found that 1 gm. of radium in equilibrium with its short-lived disintegration products develops 140 calories of heat per hour, or enough to heat more than the same mass of water from 0° C. to 100° C. In accordance with the slow transformation of radium into the end-product lead, this generation of heat gradually diminishes. In the time required for 1 gm. of radium to decay completely (roughly 20,000 years) it must evolve $3{\cdot}4 \times 10^9$ calories; for purposes of comparison it may be mentioned that we should have to burn about half a ton of coal in order to produce this quantity of heat. Even the transformation of 1 gm. of oxy-hydrogen gas into water only develops about a millionth part of the above amount of heat, viz. 3,700 calories, and it is well known that this reaction is associated with a heat of combustion which is unusually high for chemical changes.

The following consideration gives us information as to the origin of the large quantities of heat generated by radioactive substances. From the velocity and mass of the α-rays and recoil rays, and from the energy of the β- and γ-rays, which is likewise known, we can calculate the total energy liberated by the preparation in the form of rays. For complete absorption of the rays, this energy must be transformed into heat. In this way we obtain for the amount of heat resulting solely from the absorption of the rays from 1 gm. of radium, together with its disintegration products, the value 140·1 calories per hour; of this amount 124·4 calories are produced by the α-rays and the recoil rays, 6·3 calories by the β-rays, and 9·4 calories by the γ-rays. Since the quantity of heat found experimentally agrees with this value within the limits of error of experiment, we must conclude that the heat developed by radioactive substances is due to the energy of the rays, and not to any additional inter-atomic processes.

By means of sensitive apparatus it can also be proved that polonium and even thorium and uranium are sources of continuous development of heat, exactly as we should expect from the disintegration theory. One gram of uranium in equilibrium with its disintegration products generates about 9×10^{-5} calories per hour. Information on the calorimetric detection of mesothorium in radium is given on p. 56.

5. Chemical Effects

(*Note.* The study of the chemical effects of the rays from radium is sometimes called 'radio-chemistry' in the narrower sense, just as the designation 'photo-chemistry' is applied to the study of the chemical effects produced by light. Throughout this book, however, we shall confine the use of the term 'radio-chemistry' to the chemistry of the radio-elements, or the study of their chemical properties and reactions, since no other short name is available for this much greater and much more important branch of the science.)

It has already been mentioned that many similarities exist between the chemical effects produced by the rays from radium and those produced by light. In our discussion of the ionization in gases we associated it with the better-known case of electrolytic conduction. It will serve a useful purpose if, both in the individual examples and in the theory of the phenomena of the chemical effects of the rays from radium, we likewise always draw attention to the analogous photochemical effects. We shall first discuss a number of experiments on the chemical changes brought about by the action of the rays from radium.

(*a*) *Chemical Effects in Solids*

Colourless glass becomes coloured under the action of the rays from radium; ordinary sodium glass and glass containing manganese become violet, whilst glass containing iron or potassium becomes brown. The coloration is annulled by the action of light, and still more quickly by the action of heat, and at the same time the phenomena of luminescence already mentioned make their appearance (see p. 245). Precious stones and minerals also alter in colour; diamond is transformed superficially into graphite, blue sapphire becomes yellow (synthetic sapphires remain unchanged), bright green fluorspar becomes dark blue, red kunzite green, and colourless rock-salt blue or brown. Pleochroic haloes that are to be seen in many minerals (see p. 269), and which are a consequence of radioactive inclusions, show the same colorations as can be produced by artificially raying the same minerals. It is well known that many glasses assume a yellow, green, or violet colour also when they are subject to the action of light for a long period of time, or when the intensity of the radiation is large.

In the case of pleochroic haloes we have a particularly striking example of the great similarity between the action of α-rays and photochemical processes. In the vicinity of the dark rings the mica is sometimes more transparent to light than in places that have not been affected at all by α-rays [1]. Quite a similar phenomenon can be observed in photographic plates near to regions that have been strongly blackened ('Eberhard Effect').

One of the best-known photochemical effects is the bleaching of dyes. An analogous destruction of the colour by means of the rays from radium can also be detected with, say, indigo or chlorophyll. These reactions are more obvious in solutions or with strips of material soaked in the solutions than when the dye is in the compact solid form.

It is well known that light exerts a destructive action on paper containing lignin, in the presence of air. The rays from radium exert a much more energetic destructive action on every kind of paper, on silk, linen, celluloid, and the like. The action is here probably due to oxidation by the ozone produced by the rays. In a similar manner, paraffin wax, vaseline, and rubber become hard, and tap-grease is decomposed with evolution of CO_2 by the action of the rays. These phenomena must be taken into consideration in the construction of apparatus for radioactive purposes.

Yellow phosphorus is transformed into the red variety under the action of light; exactly the same effect has been observed with the rays from radium. When light is incident on selenium the electrical resistance of the selenium is found to diminish, and this fact is the basis of many processes, such as light-telephony and the ignition of explosives from a distance. The resistance of selenium also diminishes when the rays from radium are incident upon it. Nitrogen iodide explodes instantaneously when subjected to the light from burning magnesium; about 20 seconds are required before the explosion takes place when the nitrogen iodide is submitted to the α-radiation from 3 millicuries of radon at a distance of 1 cm. During this period some 10^7 or 10^8 α-particles impinge on the nitrogen iodide.

Finally, the reducing action of the rays from radium on silver chloride and silver bromide is also of great importance, and quite analogous to the action of light. It constitutes the basis of the photographic action of the rays, and for this reason it has already been mentioned on p. 240.

(*b*) *Chemical Effects in Liquids*

The decomposition of water under the influence of the rays from radium is of especial importance. In this process, gases consisting of H_2, O_2, and O_3 are liberated, and simultaneously a little hydrogen peroxide is formed in the water. We can picture the processes by means of the following two equations:

$$2H_2O = 2H_2+O_2,$$

whereby some of the O-atoms initially formed go to form ozone with the O_2, and

$$2H_2O = H_2O_2+H_2.$$

Most of the decomposed water molecules follow the first reaction, and so form oxy-hydrogen gas, but owing to the second reaction there is always an excess of hydrogen mixed with it. The amount of the decomposition products obtained from a given quantity of a dissolved salt of radium depends first on the fraction of the α-, β-, and γ-rays absorbed by the water, and thus on the dimensions of the containing vessel, and, secondly, it depends particularly on the magnitude of the gas space over the liquid, because this determines the relative distribution of the radon between the liquid and the gas phases (cf. p. 226). As an approximate estimate, it may be stated that a daily production of 20 c.c. of gas has been observed from 1 gm. of radium in solution.

This continuous evolution of appreciable quantities of oxy-hydrogen gas can give rise to much trouble when radium preparations are sealed off in glass tubes before being completely dry. The pressure of the gases developed may burst the tube, and a slight and sudden application of heat may result in a veritable explosion. For this reason it is absolutely necessary, before sealing off strong preparations, to maintain them at a temperature of over 100° C. for several hours.

If only the penetrating rays from radium are used for the decomposition of water, the velocity of decomposition is very much smaller, as when the radium preparation is enclosed in a double-walled vessel filled with water. It can be increased by enclosing the outer vessel in a sheath of lead. In this case secondary β-rays are emitted by the lead, and since they are absorbed by the water more strongly than the primary γ-rays, they give rise to a bigger effect. This experiment has been mentioned because it presents a certain formal analogy to what is called 'optical sensitization'; thus a photographic plate becomes sensitive to red rays when it contains eosin, and after the addition of chlorine to a mixture of hydrogen and oxygen the last two gases combine by virtue of those rays that are absorbed by the chlorine, whereas in the absence of chlorine the action of visible light is only a slow one.

The decomposition of water by the two reactions mentioned above can also be brought about by means of light, particularly by ultraviolet light.

It has been mentioned that a certain proportion of hydrogen peroxide is formed from water under the action of the rays from radium. The reverse reaction, the decomposition of hydrogen peroxide, is more markedly influenced by the rays. Ultra-violet light produces the same effect; in fact, a mercury lamp acts incomparably more quickly than even a strong preparation of radium.

From amongst other reactions that take place in liquids, it may be mentioned that in the mercury oxalate actinometer oxalic acid is decomposed by the rays from radium as well as by light:

$$2HgCl_2+C_2O_4(NH_4)_2 = Hg_2Cl_2+2CO_2+2NH_4Cl.$$

The amount of calomel precipitated, or the amount of carbon dioxide liberated serves as a measure of the intensity of the light. Other reactions that have been described are also in complete qualitative agreement with the transformations wrought by light, but in general they take place several hundred times more slowly than when a mercury lamp is used as the source of light. The following are examples of such reactions: the liberation of iodine from a solution of iodine in chloroform; the decomposition of liquid HBr or an aqueous solution of HBr, or of HI or KI solutions; the precipitation of silver from dissolved silver nitrate; the decomposition of nitric acid into nitrous acid, nitric oxide, and oxygen.

The observation that platinum vessels are attacked by solutions of radium chloride is explained by the liberation of nascent chlorine, which in its turn dissolves the platinum.

(c) *Chemical Effects in Gases*

The most striking of these is the formation of ozone from oxygen; the characteristic smell of ozone is detectable in the proximity of every preparation that is emitting intense radiation. In consequence of this, metals like lead, mercury, and aluminium, and also paper, linen, and the like (see above, p. 249), are oxidized in a longer or shorter interval of time when they are situated in the neighbourhood of radioactive preparations. In particular, the formation of ozone has also been accurately investigated quantitatively, but we shall defer details of this work until we discuss the theory of the chemical action of the rays (see p. 255).

Mention should be made of a few other reactions in gaseous systems that have been studied in some detail. They include the oxidation of hydrogen, carbon monoxide, and various hydrocarbons (methane, ethane, propane, and butane); the decomposition of carbon monoxide, ammonia, hydrogen chloride, hydrogen bromide, and nitrous oxide; the synthesis of water, ammonia, and hydrogen chloride; the polymerization of acetylene, cyanogen, and hydrogen cyanide. The photochemical processes corresponding to these changes are so well known that it is hardly necessary to consider them in further detail here.

Under the influence of radiation, nitrogen also becomes more reactive; a diminution in pressure due to the influence of α-rays has been observed in glass flasks filled with pure nitrogen, and having various substances (Na, K, S, P, I, As, Mg, Hg) on their inner walls.

(d) *General Nature of the Chemical Reactions produced by the Rays from Radium*

We have not classified the reactions referred to above to indicate whether they are caused by α-, β-, or γ-rays, because in many cases details on this point are not available, and in most of the cases in which the parts played by the different kinds of rays has been studied it has been found that all three produce the same changes. In general we can say that the α-rays have the strongest action, if this is not prevented by their ready absorbability. Most reactions with gases have been carried out with radon, in which case the major part of the action can certainly be ascribed to the α-rays; this also holds for the decomposition of water into oxy-hydrogen gas, which has also been established by the use of polonium, a pure α-rayer; likewise for the breaking up of nitrogen iodide. For this reason small quantities of radioactive substances can bring about colorations in glass only to a depth of a few hundredths of a millimetre, corresponding exactly to the phenomenon of pleochroic haloes (see p. 269), in which the β- and γ-rays emitted simultaneously with the α-rays also have no appreciable action.

In other reactions the β-rays are mainly responsible for the changes, for they are able to penetrate more deeply by virtue of the fact that they are corpuscular rays and may possess considerable energy. In particular, in the separation of iodine from iodoform in benzene, α-rays appear to be quite ineffective. Like ultra-violet light, β-rays decompose water into hydrogen peroxide and hydrogen, in contrast to α-rays.

The absorption of the γ-rays is so small that their quantitative effect cannot be large, owing to the small losses of energy involved (cf. p. 250 on the sensitization for γ-rays). In spite of this, they are the only rays which call for consideration from the viewpoint of deep-seated volume effects; this is the basis of their prevalent application in medicine.

From the illustrations that have been mentioned it is clear that, both in solids and in liquids and gases, the chemical actions produced by the rays from radium are extraordinarily similar to those of light, at least in their outward manifestations. Many reactions are quite identical, and this is particularly true of those reactions caused by the penetrating rays, in which case the radioactive preparation used in the experiment is enclosed in a glass tube. From the theoretical viewpoint this agreement can be readily understood, for the γ-rays are nothing else than light of very short wave-length. When the more readily absorbable rays are also admitted, the effects are very much stronger, but even then they are in all probability not different qualitatively from those produced by light. Nevertheless, reactions like the pronounced formation of ozone take place under such conditions quickly and to an appreciable extent, although they would hardly be perceptible by the use of light. It should be particularly emphasized that in the decomposition of ammonia by α-rays an alteration of the pressure or an addition of foreign gases (N_2, H_2, N_2H_4) exerts a very similar influence, even quantitatively, to that observed in the case of photochemical decomposition [2].

A further similarity with photochemistry is embodied particularly in the fact that one and the same reaction is often favoured in both directions. But whereas in the case of light, as with hydrogen bromide, it has been established that different wave-lengths produce opposite effects, such a differentiation has not hitherto been observed when the rays from radium are used. Thus radon decomposes water, and at the same time brings about the combination of oxy-hydrogen gas; an analogous behaviour is found with ammonia. It is sometimes regarded as surprising that in certain cases light produces effects the reverse of those due to radium rays, as when colorations in minerals due to the latter rays are made to disappear under the action of illumination by light. But when we remember that the effects produced by different kinds of light are often opposed to one another, we can hardly doubt the essential similarity between the processes effected by light and by the rays from radium.

Of especial importance is the fact that the temperature coefficient of the velocity of reactions actuated by the rays from radium is just as small as in photochemical reactions; for the decomposition of hydrogen peroxide it has been found to be 1·2 per 10° C.

(e) *Theory of the Chemical Reactions produced by the Rays from Radium*

Since the outward manifestations of these reactions and those of photochemistry have been found to resemble each other so closely, we may ask ourselves whether it can be assumed that the mechanism of the changes is also the same in the two cases.

The more recent physical advances on the subject of light quanta and on the connexions between radiation and matter have resulted in great progress being made in the theoretical treatment of photochemical processes. By using thermodynamical principles as a basis, the earlier tendency was to introduce a strict differentiation between those processes which take place by the addition of energy by light and those in which the light accelerates the process in a catalytic fashion. But it has now been recognized that, from this point of view, reactions which run a quite similar course have often had to be treated separately. The mechanism of the processes was first revealed by considering them from the atomistic viewpoint by means of Einstein's 'law of photochemical equivalence'. If we divide the absorbed energy of radiation by the value of Planck's element of energy corresponding to the wave-length used, then by the equivalence law we must obtain the number of elementary processes involved in the experiment. In point of fact, it is found, say when oxygen is subjected to ultra-violet radiation, that the number of elementary processes calculated in this manner is of the same order of magnitude as the number of molecules of ozone formed. To be more exact, twice as many molecules of ozone are formed, and hence we may assume that in every elementary process one O_2-molecule is split up, and the resulting $2O$ combine with two additional O_2-molecules to form $2O_3$.

It is necessary at this stage, however, to recall that, in spite of the essential validity of the law of photochemical equivalence, numerous instances of photochemical reactions are known in which the amounts actually converted are found to be smaller and sometimes greater than those which correspond to the number of the elementary processes calculated, for it is not the elementary processes themselves which are observed but only the chemical changes brought about by them. In certain circumstances a fraction of the absorbed light quanta only increases the thermal content of the substance instead of contributing to chemical change, and in this case the chemical yield must lag behind the yield calculated. On the other hand, an elementary process may be immediately succeeded by a 'chain reaction', and then the amount of chemical change may be enormous, and even more than a million times greater than would correspond to the law of equivalence. It will suffice to recall the well-known case of a mixture of chlorine and hydrogen. Each absorbed light quantum breaks up a molecule of Cl_2 into its constituent atoms; each free atom of chlorine is able to form HCl by uniting with an H-atom from an H_2-molecule and to set at liberty the other H-atom; this free H-atom can in turn react with Cl_2 to form a molecule of HCl and liberate Cl, and so the process goes on. In this way it is possible to explain why, with a mixture of equivalent amounts of hydrogen and chlorine, a single quantum of light is able to promote the formation of a very large number of molecules of HCl.

Now if we wish to examine the question as to whether the chemical reactions called forth by the rays from radium also obey the law of photochemical equivalence, we must first adequately appreciate that the number of elementary processes is determined not by the number of primary α- or β-particles fired into the reacting system, but by the number of secondary ions produced by these particles. This result emerged quite clearly from an investigation on the formation of ozone by cathode rays, which is mentioned here because the mechanism of this reaction is intimately related to that of the changes brought about by the rays from radium. Cathode rays were allowed to enter a vessel containing oxygen, and then determinations were made first of the amount of ozone formed, and, secondly, by measurement of the saturation current, of the total number of ions produced in the reaction vessel. Whereas the number of primary electrons projected into the vessel was from a hundred to a thousand times smaller than the number of molecules of ozone formed, good agreement was found between the number of ozone molecules and the number of pairs of secondary ions produced in the reaction vessel. As we have seen on p. 11, a single electron can give rise to many thousands of ions as a result of ionization by collision, and it is obviously these ions of secondary origin that give rise to an equal number of chemical elementary processes in accordance with the law of photochemical equivalence.

Reverting to the case of the rays from radium, it is necessary to apply an exactly corresponding calculation of the number of electrons taking part in the reaction. Here also it is a question of a dependent electrical conductivity caused and sustained by an outside ionizing agency, and the number of ions contributing to the conduction of electricity is much greater than the number of primary particles entering the gas, the former number sometimes being as much as 10^5 times the latter (cf. p. 27). Now the total number of pairs of ions produced by a radium preparation of definite strength can readily be found from the saturation current. In the formation of ozone from oxygen under the action of the rays from radon it was thus possible to compare the number of molecules of ozone formed with the number of pairs of ions produced, and when this was done it was found that the requirement of equivalence was excellently confirmed within the limits of experimental error. Here also we should obtain much too small a value for the number of elementary chemical processes if we were to regard the charges carried by the primary α-particles fired into the gas as being the determining factor in the reaction, instead of those of the pairs of secondary ions formed.

This equivalence between the number of molecules entering into chemical reaction and the total number of ions formed by the α-rays has been proved not only for the formation of ozone, but also in the formation of water, ammonia, and hydrogen chloride from the constituent elements, and also in the decomposition of ammonia, hydrogen chloride, hydrogen bromide, and carbon monoxide. Moreover, it has also been established in liquid systems, as in the decomposition of water, liquid hydrogen bromide, and in an acid aqueous solution of potassium iodide. In accordance with this equivalence it has been possible to show that a definite amount of chemical change is associated with the presence of a definite number of ions, in a similar manner as in Faraday's law. In contradistinction from Faraday's law, however, these ions do not participate in the transport of current, and hence in this case we generally speak only of an 'iono-chemical equivalence'.

Chemical actions are here brought about in a roundabout way by the formation of excited atoms or of ions. The recognition of this fact renders it intelligible why the utilization of the radiative energy in its transformation into chemical energy is such a poor one, both with light and with the rays from radium, assuming the validity of the simple law of equivalence, but not in the case of chain reactions. On the assumption that the first step in the process is complete ionization, $5{\cdot}5\times 10^{-11}$ ergs must be involved in the formation of a pair of ions in air. Now if one such pair of ions induces only one molecule to take part in chemical change, the greatest known heat of reaction in such a transformation, which we shall regard as a measure of the free energy, amounts to no more than 7×10^{-12} ergs. Hence only about 13 per cent. of the energy can be stored up in the form of chemical energy. Moreover, since usually more than one pair of ions per molecule is necessary, and the heat of reaction is smaller than the above amount, it follows that the utilization of the energy is in general still worse (2 to 3 per cent.).†

It is particularly noteworthy that the iono-chemical law of equivalence also holds when the chemical reaction takes place in the direction of the free energy, so that here also a much greater radiative than chemical energy is involved. It makes absolutely no difference in the number of ions required that now no chemical energy whatsoever must be stored up; the large thermodynamic difference does not make itself manifest, and from this we must conclude that even in such cases we may not conceive the action of the rays to be of a catalytic nature (cf. what was said above on p. 253 about the inadvisability of the older division into two categories). The experiments supply unequivocal evidence that the effectiveness, say of radon, is of the same order for a reaction which proceeds in the direction of its own free chemical energy, as for one that proceeds in the reverse direction. This observation is quite similar to that which has been made in the field of photochemistry. Thus in the decomposition of ammonia by means of ultra-violet light it has been established that the same intensity of light is necessary for the decomposition of a given quantity of ammonia, no matter on which side of thermodynamic equilibrium we may be.

Finally, work carried out in recent years has revealed that the same kind of chain reactions can be initiated by the rays from radium as by light. In the case of an equivalent mixture of hydrogen and chlorine it was possible to show that ionization by α-rays gives rise to chains of the same length (a few thousand molecules) as photochemical excitation, and that the temperature coefficient of the two reactions is the same. The synthesis of phosgene from CO and Cl_2 also gives the same yield when the absorbed light quanta are replaced by an equal number of pairs of ions produced by α-rays. Further examples of chain reactions that can be initiated by the radiations from radium are the chlorination

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† The calculation comes out more favourably when we do not assume complete ionization as a preliminary to the change, but rather only excitation of an electron.

of benzene, and the oxidation by oxygen of sodium sulphite dissolved in water. This last reaction is noteworthy in that it takes place in a liquid system.

From what has been said there can be no doubt that the chemical reactions that take place under the influence of the rays from radium can be interpreted theoretically by the application of the ideas developed in photochemistry. But at the present time the development of a detailed reaction mechanism is faced with considerable difficulties [3].

6. Colloido-Chemical Effects

In this section we shall discuss individual effects produced by the rays from radium, in so far as they are intimately related to questions of colloidal chemistry. It is, of course, not possible to delineate them sharply from the topics already treated. Thus it is known that the photographic process, of which we have previously spoken, must also be considered from the colloido-chemical viewpoint, and the colorations dealt with on p. 245 (second footnote) likewise present certain colloido-chemical problems for solution.

The study of the action of the rays from radium on the stability of colloidal solutions belongs to colloido-chemical investigations in the narrower sense. Here the interesting result has been established that positive colloids are precipitated, whereas negative ones survive the radiation. The experimental arrangement was so chosen that almost exclusively β-rays were effective, and since these carry negative charges, we can understand why they deposit the positive colloids. Thus if we add to a colloidal solution of iron hydroxide a quantity of a salt solution, say sodium acetate, just insufficient to produce a change, we can obtain turbidity of the colloid by means of radium rays, whereas a control sample that has not been subjected to radiation will remain unchanged. With a sol of cerium hydroxide it is not even necessary to sensitize it previously by means of an electrolyte; by subjecting the sol to radiation it is transformed into a coherent gel even after a period of 24 hours, whereas it remains unchanged for years when not submitted to the action of radiation.

None of the electro-negative colloids of gold, molybdenum blue (an oxide of molybdenum containing water), and vanadium pentoxide, the last of which tends to form a gel just like cerium hydroxide, were affected when subjected to radiation. Albumin, which also forms electro-negative colloidal solutions, is deposited in flocculent masses, but it is probable that this is not simply a phenomenon of discharge, but rather due to the fact that the albumin first loses its original nature.

Under certain conditions the rays from radium are also able to bring about the formation of cloud deposition. Thus if we introduce into a closed vessel distilled water, a piece of sulphur, and radon, the air space becomes filled with a persistent cloudy suspension. Since we can detect traces of sulphuric acid in the water, the explanation of the phenomenon undoubtedly seems to be that SO_2-molecules, and from these SO_3-molecules, are produced by the action of the rays, and these serve as condensation nuclei for water vapour. Visible fumes must be formed in exactly the same way as when, say, hydrogen chloride escapes into the moist air of a room. When sulphur has not been introduced into the vessel, only a very much weaker cloudiness is obtained. It is probable that in this case the nitrogen oxides formed by the action of the rays from the radon give rise to just sufficient condensation nuclei to enable a cloud to form, for if we replace the air by carbon dioxide, we can no longer observe any permanent cloud.

If we use super-saturated water vapour, condensation occurs directly on the ions formed, without the necessity for chemical reactions. This method is utilized to render visible the tracks of the rays from radioactive substances (see p. 19).

The following observation is also more or less closely related to the facts already described. By means of β-rays it is possible to increase the rate of crystallization of supercooled sulphur. If only a fraction of the sulphur drops are rayed, many more crystallization nuclei will be found among these drops than among those that have not been so treated.

7. Biological and Therapeutic Effects

Not long after the discovery of radium it was observed that this substance must not be handled without precaution. If we allow a strong radium preparation to lie for some time on the skin, sores develop, which are very similar in appearance to burns. Whereas in the case of the action of heat, however, our attention is immediately drawn to the danger by the resulting pain, the rays from radium have the sinister property that they do not at first give rise to any unusual feeling; with a strong preparation it is not until several days or a week or two later that the inflammatory process begins. Particularly in the earlier days of radioactive research, when the danger had not yet been recognized, many radiologists suffered from such sores on their fingers. In some cases a marked atrophy of the affected finger-tips developed, and there is a danger of cancer developing later, just as in the case of burns produced by X-rays.

The maximum daily dosage of γ-rays that a human being can tolerate without apparent harm is 0·1 röntgen unit (cf. p. 57). When working with radioactive preparations, it is advisable to carry in the pocket a miniature ionization chamber about the size of a fountain-pen and charged to a potential of a few hundred volts. By testing the extent to which discharge has taken place in the ionization chamber after the lapse of a few days, it is an easy matter to ascertain whether or not the daily dose of γ-rays to which the body has been subjected has exceeded $0{\cdot}1\,r$ unit per day.

After it had been established that the rays from radium have a physiological influence, it was suggested that they might have an application in medicine, and as a matter of fact it is possible to produce satisfactory improvement in various skin diseases, whereas healthy skin is damaged by the rays. The penetrating rays often have a healing or at least a favourable and soothing effect on internal diseases, particularly in cases of carcinoma. In most cases normal tissue is from four to seven times more resistant to the effect of the rays than diseased tissue. Moreover, much success has been attained in cases of gout and rheumatism, and science has recently been inclined to attribute the healing action of many natural springs to their content of radon. When baths are taken in water containing radon, the process involved is probably that the radon is given off by the water, and inhaled by the patient (see p. 226). A decrease in the leucocyte content of the blood has been observed as a result of the injection of radioactive solutions, and it may be possible to make use of this fact in cases of leucocytosis.

The underlying processes are not only of great importance in medical therapy—on which subject further information can be obtained from the extensive literature of medical radiology—but they are also of undoubted interest from the viewpoint of theoretical biology, in which connexion they have frequently been investigated. We can discuss here only the most important of the experimental results.

Stimulants which are harmful to an organism when administered in large doses often have a favourable influence when used in small doses. Here we may recall the fatal action of large quantities of many compounds used in medicine. Now this general rule, which has occasionally been designated a 'fundamental law of biology', has been completely vindicated in its application to the influence of the rays from radium. Young plants exposed to the rays from radium are found to exhibit a distinct improvement in their growth when only small doses are used, whereas large doses are highly detrimental to the development of the plants, as can be seen from Fig. 51 (Plate V). By the action of a sufficiently strong source of radiation we can destroy the vitality of seeds, and kill bacteria (cholera, typhus, anthrax, streptococcus) and also higher animals such as caterpillars. Germ-cells are especially sensitive; if we subject ova or spermatozoa to the action of the rays, we can observe different kinds of developmental arrest. Changes in the cells which cannot be detected even with the highest magnifications manifest themselves in the further development by the appearance of deviations from the normal development. A former contention of the specific action of the rays on biochemical compounds *in vitro* has not been verified. On the other hand, β-rays have a marked capacity for haemolytic action, and this fact may perhaps be made use of in connexion with their application in medical practice.

The danger of 'burning' the fingers, which is considerable where radium preparations are often handled, can be diminished by the use of rubber finger-caps pulled over the ends of the fingers which actually come into contact with the tubes containing the substance. They suffice to screen off the soft β-rays, which appear to be mainly responsible for the burns, and they are to be preferred to harder material, because in the latter case secondary β-rays are likely to be produced by the γ-rays incident upon it. But the chief advantage resulting from their use is the absence of any hindrance to the handling of preparations with which one is working, say in transferring radioactive material from one tube to another, and similar operations. With strong preparations, however, finger-caps give inadequate protection, and suitable forceps should be used. Radium burns are particularly troublesome, because it frequently happens that even if they mend there remains a supersensitiveness of the skin, and the rays from quite a small quantity of radium are then sufficient to give rise to renewed inflammation. Here we have what is from the theoretical point of view a very interesting case of 'indirect' or 'physical anaphylaxis'; the anaphylactogens are not directly introduced into the organism as in ordinary anaphylaxis (e.g. as a foreign type of albumin), but are formed in the organism by the action of the rays.

Biologically, neutrons are roughly ten times as effective as X-rays in respect of the change they bring about in the blood picture of a rat. The maximum permissible daily dosage of neutrons is estimated to be one-tenth that for γ- or X-rays, or $0{\cdot}01\,r$ unit (cf. p. 258) [4].

An important biological effect of γ- and of X-rays is the increase produced by them in the rate of mutation of genes. All types of mutation occur, including spontaneous ones, but no types are observed which are specific to the rays. The rate of mutation is found to be independent of the wave-length of the rays and to depend only on the total amount of the incident radiation. The interpretation of these results seems to be that one direct 'hit' suffices for the initiation of mutation by X-rays and by γ-rays, and that this hit consists in the formation of a pair of ions. Various speculations have been made, according to which the cosmic rays (see p. 278) are held responsible for the production of new types, but these lack experimental support.

8. Mechanical Effects

The mechanical effects are always brought about indirectly by electrical or by chemical processes. Thus, when it was observed that glass tubes into which radium had been sealed exploded, the cause of this was to be found in the excessive pressure brought about by the development of oxy-hydrogen gas (see p. 250). The assumption of electrical discharges, which have sometimes been held responsible for this effect, appears to be quite superfluous. Quartz tubes in which strong radium preparations have been kept develop a very characteristic meshwork of fine cracks.

The 'radium clock' provides a pretty experiment for demonstration purposes, and illustrates how the rays from radium may be utilized indirectly to give rise to motion. It consists of a small glass tube containing radium, supported on an insulating quartz rod inside a somewhat wider evacuated glass tube, and having attached to its lower extremity two electroscope leaves. Since the β-rays penetrate the inner glass tube and thus continuously carry away negative charges, whereas the positively charged α-rays are retained by the tube, it follows that the latter will become more and more positively charged. In consequence of this the electroscope leaves become more and more deflected until they touch the glass walls of the outer vessel, when they are discharged and collapse, and the whole process is then repeated time and time again with the regularity of a 'clock'. The charge collected on the inner glass tube in such an arrangement may suffice to raise it to a potential of several hundred thousand volts.

The phenomenon of the 'ionic wind' is of great theoretical interest. The α-rays show this effect best, but β- and γ-rays are also effective. When the air between the plates of an ionization chamber is ionized, then on applying an electric field the air between the plates is set in motion. The direction of this current of air is from regions of stronger to regions of weaker ionization. It is caused by the fact that the moving ions exert a viscous drag on the surrounding molecules of air. Even when the distribution of ionization in the chamber is uniform, an ionic wind can still be detected, because the dragging action of the positive ions is somewhat greater than that due to the negative ions.

9. Atomic Disruption

The phenomenon of atomic disruption is the most potent action caused by the rays from radio-elements, and has as its basis the fact that we are enabled in this way to bombard individual atoms by means of rays which have a very high localized energy content. Atomic disruption has recently also been achieved by the aid of highly energetic protons and deuterons, and by neutrons even of small energy, and the results of such work, as well as those obtained with the rays from radium, are dealt with elsewhere in this book (see Chapter X, p. 108). It will suffice here if we again draw attention to this theoretically most important application of the sources of energy rendered available by the rays from radium, owing to its significant bearing on our ideas of the structure of atoms.

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XXV

THE SIGNIFICANCE OF RADIOACTIVITY IN GEOLOGY AND IN COSMICAL CHEMISTRY AND PHYSICS

1. Determination of the Age of Minerals

The velocities of radioactive decay are natural constants characteristic of the disintegration processes under consideration, and are independent of all external conditions. They can be utilized for the measurement of time, for since they supply us with the functional relation between the amount of the substance transformed and the time, it is clear that the velocity of decay enables us to calculate the latter quantity when the former is known. Thus we could readily recognize the lapse of an hour without the aid of a clock if we noted the time taken for the activity of a preparation of ThC to fall to 49 per cent. of its initial value, as measured by a method, say, of steady deflexion. Such methods for the measurement of time have actually attained great importance in geology, the most significant of them being the following:

(*a*) Determination of the uranium-lead content of uranium minerals.
(*b*) Determination of the helium content of minerals and rocks.
(*c*) Measurement of the intensity of coloration of pleochroic haloes.

In all three cases we can readily calculate the time necessary for the attainment of the values found.

(*a*) *Age Determination from the Uranium-lead Content*

We have already seen (cf. Table 34, p. 143) that uranium, during its successive disintegrations, suffers the loss of eight α-particles before it reaches the stable end-product RaG, which is a type of lead. The following expression represents this transition:

$$\underset{238}{\mathrm{U}} \longrightarrow \underset{32}{8\mathrm{He}} + \underset{206}{\mathrm{RaG}}.\dagger$$

From this, and from the disintegration constant of uranium ($\lambda = 1{\cdot}57 \times 10^{-10}$ year^{-1}) it follows that 1 gm. of uranium produces in one year an amount of RaG equal to $1{\cdot}57 \times 10^{-10} \times \frac{206}{238{\cdot}07} = 1{\cdot}36 \times 10^{-10}$ gm. Thus if a mineral contains $1{\cdot}36 \times 10^{-2}$ gm. RaG to every 1 gm. of uranium, it follows that the age of the mineral is 100 million years. In general,

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† In the considerations of this chapter we shall disregard the influence of actino-uranium (^{235}U) on the results. This is permissible, seeing that ^{235}U constitutes less than 1 per cent. by weight of the element uranium (see p. 221), and hence, in spite of its more rapid rate of decay, the effect of disregarding it does not exceed the limits of error of the age determinations.

the age of a mineral in years is found by this method by dividing the amount of RaG corresponding to 1 gm. uranium by $1{\cdot}36\times10^{-10}$, or by multiplying it by $7{\cdot}4\times10^{9} = 7{,}400$ millions. In this way we obtain the following useful formula for an approximate estimate of the age:

$$\text{Age} = \frac{\text{RaG}}{\text{U}}\times 7{,}400 \text{ million years;}$$

where RaG is the uranium-lead content, and U the uranium content of the mineral concerned. In this calculation no account has been taken of the fact that in the course of long periods of time the quantity of uranium will have appreciably diminished as a result of disintegration, and that a somewhat lesser time will have been sufficient to produce the amount of RaG found, from the previously greater quantity of uranium. To a first approximation we do justice to this correction by utilizing the following formula for the determination of the age of the mineral:

$$\text{Age} = \frac{\text{RaG}}{\text{U}+0{\cdot}58\text{RaG}}\times 7{,}400 \text{ million years.}$$

Thus for the uranium pitchblende from Morogoro (Tanganyika Territory) the ratio RaG : U = 0·093 was obtained, whence it follows that the age of this mineral is about 600 million years. In a similar way, the age of Norwegian bröggerite, for which the ratio RaG : U = 0·127, works out to be 870 million years.

Whereas the Morogoro ore contains no ordinary lead, most uranium ores have become more or less contaminated by ordinary lead in the process of geological changes. In making use of the analysis of the mineral, we must be able to differentiate between this disturbing lead content and that of RaG, and this necessitates the performance of a determination of the combining weight of the mixture of lead isotopes (RaG+Pb) concerned, from which the Pb and RaG contents of the mineral can immediately be calculated. Thus the mixture of lead isotopes extracted from uraninite from North Carolina is found to have a combining weight of 206·4, from which it follows that 70 per cent. of the atoms of this mixed element are composed of RaG.†

The stable end-product of thorium (ThD) is also an isotope of lead, and if the mineral contains thorium, the thorium and thorium-lead contents must also be taken into consideration in the determination of the age. The simpler formula given above for the calculation of the age then assumes the following form:

$$\text{Age} = \frac{\text{RaG}+\text{ThD}}{\text{U}+0{\cdot}33\text{Th}}\times 7{,}400 \text{ million years.}$$

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† $206{\cdot}4 = 0{\cdot}7\times206+0{\cdot}3\times207{\cdot}21$, where 206 is the atomic weight of RaG, and 207·21 the combining weight of ordinary lead.

But the number of uranium minerals practically free from thorium is so considerable that the thorium content can often be disregarded, the more so because, owing to its greater half-value period, thorium is not so effective as uranium in the production of lead (factor = 0·33).

From amongst the minerals whose ages have been determined by the method described, a uraninite from the Upper Cretaceous has been found to be one of the youngest (60 million years). This number thus gives us the age of the Upper Cretaceous System, if we make what appears to be quite a plausible assumption, that the uraninite concerned is a primary mineral, and has not gained access into the formation in the course of a subsequent epoch, in which case it would be secondary. The highest reliable values that have been obtained hitherto by the lead method amount to 1,100–1,400 million years; they were established for uraninites from the Middle and Lower Pre-Cambrian.

On the assumption (cf. p. 235) that the lead isotope ThD, the end-product of the thorium series, is completely stable, we should of course be able to determine the age of thorium minerals from their lead content in a manner exactly analogous to that used for uranium minerals. Here we must eventually apply the corresponding simple correction for the amounts of admixed uranium and uranium-lead (RaG). It has been found, however, that the ages of many thorium minerals calculated in this manner are so small (in some cases scarcely 10 million years), that here sources of error of one kind or another must be involved. In most cases weathering of the metamict thorium mineral and leaching out of part of the lead are the probable causes of the too small value of the lead-thorium ratio. All in all, the correct application of the methods described to the determination of the age of geological formations always entails the careful selection of well-preserved minerals, together with proof that they are of primary origin.

A still more important limitation of the 'lead method' is based on the fact that it is only applicable to radioactive minerals in the narrower sense, which contain uranium or thorium as an essential constituent, but not to ordinary minerals and rocks. In these last the quantity of uranium amounts only to from 10^{-8} to 10^{-5} gm./gm., and of thorium from 10^{-7} to 10^{-5} gm./gm. Since, in accordance with the age of the rock, the quantity of uranium-lead and thorium-lead formed from them is still smaller by one or two powers of ten, whereas the amount of ordinary lead admixed in rocks is of the order of magnitude of 10^{-5} gm./gm., it is clearly impossible to establish by atomic-weight determinations how much uranium-lead or thorium-lead is present. (Cf., on the other hand, the 'helium method', p. 267.)

In some minerals the ratio of lead to uranium has been altered by leaching-out processes in the course of the long periods of time involved, and this constitutes one of the uncertain factors in connexion with the lead method. For this reason there is, in principle, some justification for preferring a variant of the lead method, in which the determination of the age of the mineral is based on its relative content of actinium lead (AcD) and of uranium lead (RaG). Since actino-uranium (^{235}U) disintegrates about six times as quickly as uranium I, the ratio of the amounts of AcD and RaG formed depends on the time t during which the radioactive decay of the mineral has been taking place, in accordance with the formula:

$$\frac{\mathrm{AcD}}{\mathrm{RaG}} = \frac{\text{Activity due to AcU}}{\text{Activity due to U}_\mathrm{I}} \times \frac{\lambda_{(\mathrm{U_I})}}{\lambda_{(\mathrm{AcU})}} \times \frac{(e^{\lambda_{(\mathrm{AcU})}\cdot t}-1)}{(e^{\lambda_{(\mathrm{U_I})}\cdot t}-1)}.$$

Since AcD and RaG are isotopes, we can be certain that their ratio cannot have been altered by leaching-out processes and the like, and this ratio can be determined by densitometer measurements of the hyperfine structure of the spectral line 3,572 A.U. [1].

Experiments so far carried out have shown that this method of age determination is practicable. Unfortunately several grams of lead are necessary for this purpose, and its application is therefore limited to radioactive minerals. The method cannot be used for the determination of the age of rocks.

(*b*) *Age Determination from the Helium Content*

As mentioned above, the uranium atom undergoes eight α-transformations before it is converted into RaG. The majority of these α-particles are brought to rest within the mineral, and are retained in it as gaseous helium, which does not escape until the powdered mineral sample is dissolved, fused, or heated, when it can be collected and measured. From the disintegration constant of uranium it can be calculated that 1 gm. of uranium in equilibrium with its succeeding products produces annually $1{\cdot}18\times10^{-7}$ c.c. helium. The required age in years of a uranium mineral is thus found by dividing the quantity of helium (measured in c.c.) produced by 1 gm. of uranium in the mineral by $1{\cdot}18\times10^{-7}$, or by multiplying it by $8{\cdot}5\times10^{6}$; in this way we obtain the formula

$$\text{Age} = \frac{\mathrm{He}}{\mathrm{U}} \times 8{\cdot}5 \text{ million years.}$$

In the analysis of feebly active rocks, with which we are mainly concerned here, it is usual to calculate the uranium content from the radium content on the basis of the relation that about 3×10^{6} gm. of uranium correspond in radioactive equilibrium to 1 gm. of radium (see

p. 206). For this reason it is advisable to insert the radium content directly in the above formula, which then assumes the form:

$$\text{Age} = 2{\cdot}8\frac{\text{He}}{\text{Ra}}\ \text{years.}$$

In these expressions the disintegration of the uranium has not yet been taken into account, as a result of which the values of the age come out somewhat too large (cf. what has been said above in connexion with the lead method). For the exact calculation of the age of 'old' rocks the following formula allows for the disintegration of the uranium:

$$\text{Age} = 1{\cdot}47\times 10^{10}\log\left(1+4{\cdot}42\times 10^{-10}\frac{\text{He}}{\text{Ra}}\right)\ \text{years.}$$

When thorium as well as uranium is present in the mineral, it is of course necessary to make allowance for its contribution to the production of helium. Since the helium method has recently been used frequently for rocks, and these always contain thorium as well as uranium, it will be convenient to give also a formula for calculating ages up to several $\times 10^8$ years, when the helium is derived both from thorium and uranium:

$$\text{Age} = \frac{\text{He}}{0{\cdot}35\,\text{Ra}+3{\cdot}0\times 10^{-8}\,\text{Th}}\ \text{years.}$$

When the 'helium method' was first applied to minerals, it gave values for the age which were mostly only about one-half or one-third of the values found by the 'lead method'. The reason for this was certainly to be sought in the fact that helium readily escapes from many minerals, so that only part of the helium produced by uranium and thorium during geological epochs is available for analysis. The probability of escape of helium is greater the greater the amount of helium that has accumulated in the mineral, or the greater the activity of the mineral. Thus in thorianite, which contains up to 9 c.c. helium per gram, the pressure at 0° C. can reach values even higher than 250 atmospheres; the likelihood of a loss of helium from such a mineral is so great that we can hardly expect to obtain correct values for the age from it.

Fortunately, however, the gas-analytical method of detecting helium is so sensitive that the amounts present even in rocks can readily be determined quantitatively, and at the same time we can be certain that the whole of the helium has been freshly formed by radioactive processes since the solidification of the rock. This is in marked contrast to the case of the 'lead method', where only a part of the element lead found in rocks is freshly formed RaG or ThD, the major portion of the lead having been taken up into the mineral during its formation (see above, p. 265). Nevertheless, it has been found that even rocks that contain only about 10^{-5} c.c. helium per gram suffer an appreciable loss of helium even when they are reduced to powder, and when they are left for long periods in contact with the air they may lose up to 60 per cent. of their helium content. The losses are still greater when at any time during its geological history the rock has been heated; the helium content can be driven off in this way until less than 1 per cent. remains, more especially when we are dealing with a coarse-grained rock.

It has been found, however, that fine-grained rocks have often retained the whole of the amount of helium to be expected from their geological age and their contents in uranium and thorium. By careful choice of rocks and by careful treatment of them it is therefore possible to obtain reliable values for the age by means of the 'helium method'. The evidence for this is not based solely on the fact that the values so found are in accord with determinations that have been carried out on radioactive inclusions by the 'lead method' and with other geological criteria. The most convincing evidence is supplied by the fact that rocks taken from the same geological deposit give age determinations in very good agreement, even when their uranium and thorium contents vary. The following example of this has been taken from an investigation on the age of Keweenawan trap rocks:

$\mathrm{He} \times 10^5$ (*c.c./gm.*)	$\mathrm{Ra} \times 10^{13}$ (*gm./gm.*)	$\mathrm{Th} \times 10^6$ (*gm./gm.*)	Th:U	*Age* (10^6 *years*)
5·36	1·15	1·73	5·12	555
2·30	0·58	0·63	3·68	565

Results in like good agreement have been gained from other occurrences, so that the applicability of the 'helium method' for the determination of the geological age of rocks may be taken as proven. This opens up to this method a much greater range of material than is available to the 'lead method', which is limited to radioactive minerals, and there is good prospect that by this means it will be possible to construct a complete geological time scale, even on points of detail.

In applications of the 'helium method' to rocks it is none the less necessary for us to have information on the source of the specimens and on their treatment before the commencement of the chemical analysis. But this restriction is invalid when the method is applied to metals, for these are so impervious to helium that even on heating no helium can escape from them, so that the manner of collecting and storing it is in this case of no moment. Terrestrial metals containing a sufficient admixture of uranium or thorium do not appear to be of frequent occurrence. On the other hand, it has been possible to apply the method to iron meteorites, which contain measurable amounts of uranium and helium, but their thorium content is so small that it has not yet been possible to take account of it in the analyses, and the values found for the age are therefore maximum values, which may be about 30 per cent. too high. They vary between the limits 100 and 2,800 million years, and in no case that has so far come to our notice does the age exceed that which is assumed for the planets, on the basis of various astronomical evidence. This result lends support to the assumption that the meteorites belong to the solar system, which is probable also for other reasons.

(c) *Age Determination from the Intensity of Coloration of Pleochroic Haloes*

The α-rays produce coloration in glass, quartz, mica, and in similar materials (see p. 248), the depth of coloration depending on the number of α-particles that traverse the material. A strong preparation of radium produces an appreciable coloration in the course of only a few hours, but the same effect could be produced if a preparation of one-millionth the strength were allowed to act for a period a million times longer. It follows that we can construct an empirical scale of colorations, produced, say in mica, by different amounts of radium in the course of a day's exposure. By means of this scale we can find a rough estimate of the time that would be necessary for an active substance of known strength to act on the mica in order to produce a given depth of coloration.

Such colorations actually occur in natural minerals. Thus we are familiar with varieties of the following minerals that have been coloured by means of α-rays: rock-salt, fluorspar, spinel, garnet, tourmaline, mica, and hornblende. α-particles are emitted radially from the small radioactive inclusions in such minerals, and produce coloration within a sphere of radius equal to the range of the particles. In thin sections these colorations appear as circular patches, and when examined in polarized light they show the property of 'pleochroism'—whence the name 'pleochroic haloes'. The radius of these spherical colorations amounts to only 0·0034 cm., corresponding to the range in mica of the longest α-particles here involved, those of RaC′.

The exceedingly long interval of time during which these minerals containing inclusions have been subjected to radioactive radiations makes it possible in such cases to detect the effect of an amount of radium as small as 10^{-19} gm. This amount is a million times smaller than the amount determinable by electrical means (cf. p. 225). In age determinations we must, of course, also know the amount of radioactive material contained in the inclusion, in addition to the depth of coloration. This cannot be determined directly, but from a knowledge of the mineral of which the inclusion is composed, and by measuring its size with a microscope, an estimate of the quantity of radioactive matter in the inclusion can be formed, but only very approximately. Moreover, since the intensity of coloration may have been changed during geological time by thermal and similar processes, and a reversal may have occurred by over-exposure to the effect of the rays, just like the weakening (reversal) of the image which occurs with a photographic plate, this method of determining the age of minerals cannot lay great claims to reliability.

When attention is paid to details of the coloration, the phenomenon of pleochroic haloes can be made the basis of a better method for determining the age of minerals. The uranium mineral, which as an inclusion forms the centre of a halo, contains all the α-rayers of the uranium and actinium disintegration series in radioactive equilibrium. The various α-rays have different ranges, and the intensity of the resulting coloration appears to be greatest towards the end of the range. (Cf. the ionizing action of the rays, Fig. 11, p. 28. However, little more is known about the nature of the coloration process than that it consists of a kind of photochemical reaction, and accordingly the coloration also varies with the chemical composition of the particular variety of mica—cf. p. 248.) An especially striking similarity to the action of light on a photographic plate consists in the fact that close to intense regions of blackening there are developed zones of particular clearness ('Eberhard effect', cf. p. 248). All this has the result that the ranges of the various α-rayers manifest themselves in the microscopic image of a pleochroic halo in biotite mica as separate rings (see Fig. 52, Plate IV) [2]. By special technical means (halo photometer) it is possible to record photometrically the intensity of blackening along the diameter of a halo. Fig. 53 shows the curve of blackening corresponding to the halo in Fig. 52, and Fig. 54 gives a composite tracing of the records for two normal haloes (uranium). In this last figure the kinks in the curve are designated by letters, and the rings in the halo which can be detected optically are indicated below the figure. The distances of these kinks from the centre of the halo can be accurately measured, and by taking account of the chemical composition of the material, the ranges in air can be calculated from them (range in mica $= c.\ 5\times 10^{-4}$ of the range in air). For the kinks in the curve designated by the letters A to J the average values (Table 50) of the ranges were found.

From this table we see that the point A agrees with the end of the range of the α-particles from RaC′; D with RaA; F with the nearly equal ranges of radon and polonium; H with Ra, Io, and $\mathrm{U_{II}}$; and I with $\mathrm{U_I}$. It is thus possible to account for all the α-rayers of the uranium-radium series.

The maxima of the curve of blackening (B, E, G, and J) have naturally no simple relation with the ranges, but the minimum C remains to be explained. This appears to correspond to the end of the ranges of the α-particles from actinon and AcC. Since the parent substance of the actinium series, actino-uranium, has a shorter half-value period than uranium I, it is to be expected that the ring corresponding to (An+AcC) will be found to stand out more strongly relative to the others, the greater the age of the halo. This has been found to be the case, and the photometric measurement of pleochroic haloes can be used, on the strength of this fact, as the basis of a method for the determination of geological time.

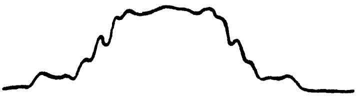

Fig. 53. Photometer Record of a Normal Halo in Biotite (Murray Bay, Quebec).

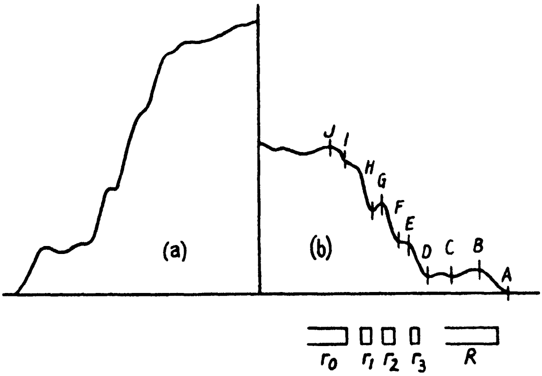

Fig. 54. Composite Photometer Record of Normal Haloes in Biotite from (a) Mt. Apatite, Maine; (b) Murray Bay, Quebec.

Table 50

Dimensions of the Halo Rings (Uranium Haloes)

	A	*B*	*C*	*D*	*E*	*F*	*G*	*H*	*I*	*J*
Radii in microns	34·4	30·5	27·1	23·1	20·6	19·7	16·9	15·6	12·4	9·5
Air equivalent in cm.	6·97	6·19	5·50	4·69	4·18	4·00	3·43	3·16	2·52	1·93

Up to the present even this improved method has not yielded such accurate results as the lead or the helium methods, and it is also far from being so generally applicable as the latter methods. But the investigations on pleochroic haloes merit attention for other reasons also. The agreement of the ranges found in them with those which can now be observed affords proof of the fact that even millions of years ago the ranges of the α-raying radioactive substances, and hence also their rates of decay (cf. p. 105), were no different from what they are to-day. A ring of radius 42·6μ, corresponding to the range of ThC′ (8·62 cm.), is occasionally found along with the rings that are due to the uranium-radium series and the actinium series. Hitherto, longer ranges have never been observed in the haloes, but shorter ones of radii 8·6μ and 5·2μ and of uncertain origin have been found. The corresponding ranges in air would be 1·74 and 1·05 cm. Whereas the latter may be due to samarium (R = 1·15 cm., see p. 235), the range of 1·74 cm. is perhaps to be attributed to an α-rayer as yet unknown, and a further study of pleochroic haloes may prove fruitful in its discovery.

Analogous results to those obtained with the uranium family have also been obtained by observations with the halo photometer of pleochroic haloes due to the thorium family in biotite mica. The radii of the five rings observed in thorium haloes satisfactorily account for all the α-particle ranges of the thorium series. The results are shown in Table 51 (cf. Table 1, p. 22). The last result in the table probably has the same origin as the range 1·05 cm. found in the examination of uranium haloes. On this assumption, the mean radius of 40 such rings was found to be 1·18 cm., with a probable error of 1 per cent. The value of the range found for thorium is of particular interest, as recent values obtained directly in a Wilson cloud chamber differ appreciably (2·59 and 2·87 cm.) [3].

TABLE 51

Dimensions of the Halo Rings (Thorium Haloes)

Radii in microns	41·80	33·5	27·77	23·86	20·00	15·2	12·38	5·9
Air equivalent in cm.	8·55	6·85	5·68	4·88	4·09	3·10	2·53	1·21
Origin	ThC′	RaC′	ThA	Tn ThC	ThX RdTh	U_{II} Io Ra	Th	Sm (?)

2. The Distribution of Radioactive Substances

It has already been pointed out in Chapter XXII, on the extraction of radioactive substances, that uranium and thorium minerals serve as sources for all the radio-elements of long life. In particular, the extraction of radium from pitchblende and from carnotite was then described. It will serve a useful purpose if we now give a summary (Table 52) of the most important uranium and thorium minerals that may be used for the extraction of radium, mesothorium, and their disintegration products.

TABLE 52

Summary of the Most Important Radioactive Minerals

I. URANIUM MINERALS

Name of mineral	*Chemical constitution*	*Content of* U_3O_8	*Most important mineralogical properties*
Pitchblende, uraninite	Uranium oxide. (UO_2 with UO_3)	60–90%	Black, rounded glassy masses, rarely crystals, greenish-grey streak.
Carnotite	Potassium uranium vanadinate. $K_2O \cdot 2UO_3 \cdot V_2O_5 + 2H_2O$	55%	Greenish-yellow crusts and nodules.
Autunite, calcium uranium mica	Calcium uranium phosphate. $CaO \cdot 2UO_3 \cdot P_2O_5 + 8H_2O$	63%	Canary-green four-cornered plates.
Chalcolite, copper-uranium mica	Copper uranium phosphate. $CuO \cdot 2UO_3 \cdot P_2O_5 + 8H_2O$	61%	Emerald-green four-cornered plates.
Uranium ochre	Various uranates and uranium sulphates	*c.* 17%	Intense yellow, earthy or in tangled threads.
Casolite	Lead uranium silicate. $3(PbO \cdot UO_3 \cdot SiO_2) + 4H_2O$	48%	Yellow, earthy.
Pilbarite	Thorium lead uranate	27%, with 31% ThO_2	Yellow, knotty.

The other uranium minerals, like bröggerite, cleveite, etc., have so far been of no economic importance.

II. THORIUM MINERALS

Name of mineral	*Chemical constitution*	*Content of* ThO_2	*Most important mineralogical properties*
Monazite	Phosphates of the Cerium Earths+Th	Up to 18%, with 31·3% CeO_2	Monoclinic.
Thorite, orangite	$ThSiO_4 + H_2O$	Up to 81·5%	Tetragonal, isomorphic with zircon, $ZrSiO_4$.
Thorianite	$(ThU)O_2$	53%, with 47% UO_2	Regular, related to crystallized uranium pitchblende.

It will be noticed from the above tables that pilbarite and thorianite are minerals intermediate between those of uranium and those of thorium.

Noteworthy is the fact that even minerals like bröggerite and thorianite, although they are very old and strongly radioactive, have been able to retain completely or in large measure the original arrangement of their crystal lattice, as has been established by means of investigations with X-rays. Nevertheless, it can be calculated that about every eighth uranium atom must have disintegrated since the bröggerite was formed, and the crystal lattice must have been traversed by about as many α-particles as there are uranium atoms contained in the lattice.

Small quantities of thorium, uranium, and of their disintegration products can be detected in rocks, in fresh- and in sea-water, in the atmosphere, and in meteorites. To establish the presence of thorium or of uranium it is customary to measure the constant amount of thoron or of radon present in the material, since these can readily be recognized.

The mean uranium content of 1 gm. of rock in the earth's crust amounts to 4×10^{-6} gm., corresponding to $1{\cdot}3\times10^{-12}$ gm. of radium, the thorium content being $1{\cdot}6\times10^{-5}$ gm., but these values vary very much with the nature of the rock. Acid rocks are much more strongly active than basic rocks. Further information on this subject is given in Table 53, which contains the uranium and thorium values for various granites, basalts, and deep-seated rocks, and in which the potassium contents of the rocks are also entered, as well as the amount of heat generated per annum by all these radioactive constituents. We shall return to the geological significance of this production of heat in the next section.

TABLE 53

Radioactive Content and Heat Production of Different Rocks

	1 gm. of rock			
	contains			
Type of rock	*Uranium* (10^{-6} *gm.*)	*Thorium* (10^{-6} *gm.*)	*Potassium* (10^{-2} *gm.*)	*yields per annum heat* (10^{-6} *cal.*)
Granite . . .	9·0	20·0	3·4	12·1
Grano-diorite . .	7·7	18·0	2·5	10·5
Diorite . . .	4·0	6·0	1·7	4·8
Central basalts:				
Continental . .	3·5	9·1	1·9	5·1
Oceanic. . .	3·6	7·1	1·8	4·7
Plateau basalt . .	2·2	5·0	0·8	3·0
Gabbro . . .	2·4	5·1	0·7	3·2
Eclogite . . .	1·0	1·8	0·4	1·3
Peridotite. . .	1·5	3·3	0·8	2·0
Dunite . . .	1·4	3·4	0·03	1·9

According to their chemical composition, stone meteorites are still more strongly basic than the ultrabasic terrestrial rocks; in good agreement with this is the fact that the average uranium content of 20 stone meteorites has been found to be $3{\cdot}6\times10^{-7}$ gm. The uranium content of iron meteorites is still smaller; on the average it amounts to only 9×10^{-8} gm., corresponding to 3×10^{-14} gm. radium. From the more commonly occurring terrestrial metals only one specimen has been accurately examined up to the present; copper from Michigan contained less than 6×10^{-9} gm. of uranium.

The radium content of sea-water is about a thousand times less than that of the earth's crust, and averages between 10^{-15} and 10^{-16} gm. per c.c. Samples taken from near the coast are in general more active than those from the high seas. In the Pacific Ocean and in the Polar Seas the radium content is smaller than 1×10^{-15} gm.; in the Atlantic Ocean the values fluctuate between $0{\cdot}3 \times 10^{-15}$ and 40×10^{-15} gm. per c.c. (this high value was observed near the Irish coast). In the Adriatic Sea the value 2×10^{-15} gm. per c.c. has been found.

The radium content of large rivers is of the same order of magnitude as that of sea-water. Much greater activities are found in a few 'radioactive' spas and thermal springs. Their greater radioactivity is usually due only to radon that has been taken up by the water in its passage through rocks of good emanating power, and containing radium; because of this, the activity is not constant. One of the most active waters is that from the Joachimstal mines, which contains 7×10^{-10} curies or 2,050 mache units per c.c. The emanation content of various springs is given in Table 49, p. 227. In the water of individual springs, however, the element radium has also been detected in strikingly high concentration. Thus spring-waters in the neighbourhood of the thermal springs at Gastein contain 10^{-13} gm. radium per c.c., or about 100 times as much as the water of the seas and rivers. A distinctly higher radium content has been found in the water of certain petroleum bore-holes; in Germany up to 5×10^{-12} gm. radium per c.c. has been found, and in the northern Caucasus there are mineral oil sols that contain up to 10^{-10} gm. radium per c.c. It is not impossible that there may be a connexion between this concentration of radium and the genesis of the petroleum oil-fields, say by the storing of barium or uranium by the organisms responsible for the formation of the petroleum. In like manner attempts have been made to explain the occurrence of sources of helium in the regions from which petroleum is obtained.

Since the soil always contains radium, emanation is constantly diffusing with the soil gases into the atmosphere, but as we ascend to higher levels above the ground the amount of the emanation and of its disintegration products steadily becomes less and less, in consequence of their decay. The concentration of emanation in the soil gases depends naturally not only on the radium content of the rocks concerned, but also on the prevailing physical conditions. In general, the gases of granite formations are richest in emanation, and those of sandy ground poorest. Rain and frost keep the emanation at lower levels, whereas the effect of strong sunlight on the ground, of winds, and of diminishing atmospheric pressure is to facilitate its escape into the free atmosphere.

The average emanation content of the ground gases amounts to 2×10^{-13} curie per c.c., and that of the free atmosphere $1{\cdot}3 \times 10^{-16}$ curie, or only one two-thousandth part of the amount. Accordingly, the atmosphere contains on the average about 1·6 atoms of emanation in 1 c.c. By taking into account the decay of the emanation, it can be calculated that 1 cm.^{2} of the earth's surface gives off roughly 4×10^{-17} curie of emanation to the atmosphere per second.

The conditions are quite different out at sea. In mid-ocean the emanation content of the air amounts to only about 1 per cent. of the mean value over land, whereas near the coasts much higher values are found, especially with a land breeze. From this it follows that the emanation content of the air over the sea is derived mainly from the land. This is consistent with the fact that the disintegration products of thoron and actinon can also be detected in the air over land surfaces, but not over the sea, since these two forms of emanation are too short-lived to be able to traverse long distances.

A small activity is also found in atmospheric precipitation. Rain and snow do not carry down with them the gaseous emanation, but they do pick up its solid disintegration products that are suspended in the atmosphere, and in this way they derive their activity.

Attempts have frequently been made to detect spectroscopically the occurrence of uranium, radium, and thorium in the stars, but the evidence is not yet unequivocal. At the same time it is almost certain that these three elements are present at least on the sun, for since the earth has been formed from the material of the sun, and we cannot assume the new production of the initial members of the radioactive disintegration series on the earth, uranium and thorium must at that time have also been represented in the solar material. Moreover, since the process of radioactive disintegration takes place in the outer layer of the sun presumably exactly as on the earth, it is rendered extremely probable that the whole of the later members of the uranium and thorium series are also available in the material of the sun.

3. Thermal Consequences of the Distribution of Radioactive Substances

Since it was established that radioactive substances occur in all terrestrial rocks, our views on the thermal development of the earth have changed accordingly in a decisive manner. Before the discovery of this source of heat it was regarded as certain that, after its separation from the sun, the earth was in the molten state, and that from this time onwards it had continuously cooled until it attained its present temperature. On this view, however, science was led to a contradiction, to wit, that according to physical calculations the time taken up by the cooling process should not have amounted to more than about 20 million years, whereas it was necessary to assume a multiple of this period for the geological development of the earth. This conviction of geologists has meanwhile acquired a complete confirmation by the age determinations mentioned in § 1 of this chapter. The very wide distribution of radioactive substances in terrestrial rocks led to the recognition of a new source of energy, of which no account had been taken by physicists in their earlier calculations. Since all radioactive substances produce heat (see p. 246), it is clear that the cooling of the earth must at least have been retarded as a result of its content of the radio-elements.

The annual production of heat by uranium and thorium with their disintegration products, and also by potassium, are shown in Table 54.

TABLE 54

Annual Production of Heat by Uranium, Thorium, and Potassium

1 gm. of uranium in equilibrium with its disintegration products gives $7{\cdot}9 \times 10^{-1}$ cal.;
1 gm. of thorium in equilibrium with its disintegration products gives $2{\cdot}3 \times 10^{-1}$ cal.;
1 gm. of potassium gives $1{\cdot}2 \times 10^{-5}$ cal.

It is seen from this summary that the production of heat per gram of potassium is by four powers of ten smaller than that of uranium and thorium, but from the geological point of view, owing to the more widespread distribution of potassium in rocks, its thermal contribution is only by one power of ten less than those of uranium and thorium. Rubidium, on the other hand, can be neglected owing to its rarity. The development of heat by various rocks containing uranium, thorium, and potassium, calculated on the basis of the above numbers, has already been given in Table 53, p. 274.

In order to be able to state with certainty anything about the total amount of heat developed by radioactive processes in the earth, it would be necessary to know not only the uranium, thorium, and potassium contents of the rocks of the earth's crust, but those of the earth as a whole. On these points, however, there are as yet no reliable data available. On the basis of chemico-geological considerations it is probable that uranium and thorium have been concentrated mainly in the lithosphere. If the assumption be made that the rocks are still radioactive only down to a depth of 16 km., and that the average activity is equal to that of the surface rocks, the heat produced in this way is already sufficient to balance completely the amount of terrestrial heat now given out into space. In other words, on the basis of this content of radioactive substances, the earth could have retained its present temperature throughout geological epochs of unlimited duration.

As a matter of fact, it is extremely improbable that the deeper crustal layers of the earth are entirely free from radioactive elements. This view is supported by our physico-chemical knowledge on the distribution of the elements between the various layers of melts, and also by the analyses of meteorites mentioned above. The uranium content of iron meteorites (see p. 274) renders it almost certain that even in the innermost core of the earth, which most likely consists chiefly of iron, uranium is still present. We must therefore discuss the question as to what results follow when the earth's content of radioactive substances is so significant that their production of heat not only suffices to maintain the continuous flow of heat from the earth's interior to the surface and beyond into space, but also brings about a heating of the interior of the earth. Geologists have drawn attention to the fact that just such an assumption would suffice to explain many hitherto puzzling facts of the earth's history, such as geological cycles, mountain formations, and perhaps also the displacement of the continents. The excess of heat would result in a fusion of deep-seated layers and this would necessarily call forth powerful movements in the earth's interior. These movements would then again create the possibility of cooling and freezing of the layers concerned, until after millions of years fusion would again take place in a new cycle. Owing to the uncertainty of all the assumptions on the quantity and distribution of the radioactive substances in the earth, these considerations are for the time being strongly hypothetical, and for this reason we shall not discuss them in greater detail.

4. Ionization of the Atmosphere. Cosmic Radiation

It has long been known that atmospheric air is always ionized to a certain degree, and to this is due its ability to discharge electrically charged bodies (cf. p. 7). Owing to their tendency to recombination, and also because of the electrical field of the earth, these atmospheric ions would necessarily vanish in a few minutes, if provision were not continuously made for their formation afresh by one or another kind of ionizing agency.

The presence of radioactive substances in the earth and to some extent also in the atmosphere itself suggests the idea of regarding the ionization of the air as a consequence of the rays emitted by these substances. From the quantity of ions present on the average near to the ground in the atmosphere, and from the velocity of their recombination, it is necessary to conclude that 10 to 13 pairs of ions are freshly formed per second per c.c. The question now arises whether the quantity and distribution of the radioactive substances suffice for the explanation of this ionization.

In the study of this question a very curious phenomenon presented itself. Although the amount of the radio-elements in the atmosphere continuously diminishes with increasing distance from the ground (see p. 275 et seq.), and the effect of the gamma radiation emitted by the earth in the same direction must also diminish, it was found in balloon flights that the ionization produced in gamma-ray electroscopes increases again in the higher levels of the atmosphere. The explanation could only be that a penetrating radiation not of terrestrial origin is present, which, owing to the smaller absorption by the earth's atmosphere, will, of course, manifest itself more strongly at greater heights. Further investigations have shown that this radiation differs not only in its origin but also in its large amount of energy from all types of radiation known to be due to the atomic disintegration of radioactive substances.

The flux of energy of this ultra- or cosmic radiation striking the earth's surface per second is by no means insignificant, but amounts to $3{\cdot}5 \times 10^{-3}$ erg per sq. cm. It is thus equivalent to the stream of energy of several hundred α-particles per sq. cm. per second, and of about the same magnitude as the luminous and thermal energy that reaches the outer confines of the atmosphere from all the fixed stars.

With sensitive apparatus it has been possible to detect this abnormally penetrating radiation also at sea-level, where it effects the formation in the atmosphere of about 1·5 pairs of ions per c.c. per second (cf. p. 282). By the use of self-registering electrometers sent up into the atmosphere with sounding balloons, the variation of the ionizing effect of this penetrating radiation has been measured up to heights of about 25 km. At this height the pressure of the atmosphere is only 22 mm. of mercury, so that it is only necessary to take into account 3 per cent. of the atmosphere as the absorbing medium above the ionization chamber. At this level in the atmosphere the ionization rises to the value of $330I$.[†] Conversely, when self-registering electrometers were sunk in lakes, the ionizing effect of this radiation diminished continuously, and at a depth of 250 metres it amounted to less than $0{\cdot}01I$, or less than one-thirty-thousandth part of its value in the uppermost layers of the stratosphere. These experiments show that this radiation penetrates through the atmosphere from outside, and that it requires a layer of 250 m. of water to render its effect practically negligible, so that it possesses a much greater hardness than the hardest γ-rays known to us.

It has been possible to obtain more exact information on the direction of the rays by using two Geiger counters arranged some distance apart, instead of an ionization chamber, for the detection of the rays. When one of the counters is arranged vertically over the other counter, the

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[†] $1I$ = 1 pair of ions per c.c. per second.

two counters give simultaneous registrations more frequently than for other relative positions of the counters. The simultaneous functioning of the two counters is obviously brought about by penetrating particles that shoot through both counters, and the fact that this phenomenon arises more frequently when the counters are arranged one vertically over the other proves that this kind of radiation approaches the earth mainly in a vertical direction. Insertion of sheets of lead between the counters only slightly diminishes the number of simultaneous discharges (coincidences), and this proves that we are here confronted with an extraordinarily penetrating kind of radiation, and thus manifestly with cosmic rays. The fact that the rays preponderate in the vertical direction is most simply explained on the assumption that the radiation that is incident on the earth's atmosphere is more strongly absorbed in all other directions on account of the greater thickness of the atmosphere then traversed.

Although the major part of the radiation is constant at a given place, slight fluctuations have been detected, and these seem to be related to the time of day and year, and to irregularities in the earth's magnetism. On the other hand, it has been found that it shows clearly a dependence on the geographical latitude of the place of observation. When different places at sea-level are compared it is found that the intensity of the radiation is smallest at the equator, that it rises in value north and south of the equator, and reaches a 14 per cent. higher value at a latitude of 50°. This last value is maintained from there to the poles. At a height above sea-level of 4,000 m. the percentage increase in intensity with geographical latitude amounts to over 30 per cent. This dependence on latitude renders it extremely probable that an essential part of the radiation consists of charged particles.

The best experimental tool in the study of the nature of ionizing particles is the Wilson expansion chamber (see p. 19). The application of this method to the penetrating radiation was rendered more difficult by the fact that, owing to the small absolute intensity of the radiation at sea-level, cloud tracks which could be ascribed to the cosmic radiation were to be found naturally on only a small fraction of the Wilson photographs. But by combining the Wilson chamber with the above-mentioned device of the dual Geiger counter, it was possible by means of a self-releasing mechanism to effect the working of the Wilson chamber always only at the moment when a penetrating particle had traversed both counters and the Wilson chamber. By this means the number of successful exposures was more numerous, and the study of the plates obtained in this way was exceedingly fruitful. It was found that the penetrating radiation consists of approximately equal quantities of positive and of negative electrons (positrons and electrons), possessing exceedingly high energy values of 10^8 to 10^9 electron volts, and in individual cases as high as 10^{10} e.V.

This explains the results on the geographical distribution of the penetrating radiation. Theoretical considerations closely related to the explanation of why the aurora only occurs in the neighbourhood of the poles (cf. p. 283) lead us to expect that positive and negative electrons that are incident on the atmosphere from outside must possess an energy greater than 10,000 million electron volts in order that they may strike the earth at the equator. At a latitude of 30° even less than 7,000 million electron volts and at a latitude of 45° 4,000 million electron volts suffice for them to reach the earth. Round the poles north and south of latitudes 45° still smaller energies should suffice, if account had to be taken only of the magnetic field of the earth; but since a smaller energy is insufficient for the rays to penetrate the atmosphere, no further increase in the intensity is to be expected beyond these latitudes, and as already mentioned no further increase has been observed. In order to understand why the rays are able to penetrate to a depth of 250 m. in water, we must assume that some of the electrons and positrons possess an energy of more than 100,000 million electron volts.

The nature of the true 'cosmic' radiation which enters the outer atmosphere from beyond need not necessarily be the same as that observed at sea-level. Thus it has been found that at a height of 2,500 m. the intensity of the radiation coming from the west is 15 per cent. higher than that of the radiation coming from the east; this seems to indicate that in this case most of the incident particles consist of positrons. Apart from the tracks of individual particles, however, the photographs obtained with a Wilson cloud chamber also show the sudden and simultaneous appearance of a large number of tracks, all of which originate near one point within the Wilson chamber. From their position and the direction of their magnetic deflexion, these tracks seem to be due to electrons and positrons. The interpretation of these 'showers' is probably that under the influence of the extraordinarily energetic cosmic radiation, when an atom is struck it is caused to disintegrate explosively, and thus gives rise to new tracks, but the more detailed mechanism of the production of showers has not yet been elucidated. Other penetrating cosmic-ray particles have also been detected. Lighter than protons, these 'heavy electrons' are probably produced in the upper atmosphere.

Still less is known about the mode of origin of the cosmic rays than about their nature. Their practical uniformity by day and night shows that the sun is not the principal source responsible for the rays, and since no connexion with sidereal time is observable, the Milky Way is likewise excluded as the place of origin of the rays. Neither can the interior of the stars be held responsible, for the energy of the radiation would be reduced in its passage through the material of the stars. Thus the only sources that call for consideration in the production of the radiation seem to be the outer layers of the stars, nebulae, or the interstellar matter present in space. But at the present time we know of no process of transformation of matter in which energies of 10,000 million e.V. and more could be produced. Not even the hypothesis of the total annihilation of matter seems adequate for the purpose, for the complete disappearance of the mass of a proton supplies only 1,000 million e.V., and we can hardly conceive the possibility that heavier atomic nuclei vanish and in so doing transfer the whole of their energy to a positive or to a negative electron. For this reason, recourse has even been made to the conception that the cosmic radiation was formed at a much earlier period in the development of the universe, when processes now unknown to us were still possible, and that the radiation then produced has circulated uninterruptedly since that time in the universe, which according to some modern theories is a closed system. Owing to the exceedingly rarified distribution of matter in the universe, a ray that had traversed the entire universe would have experienced only about one-hundredth part of the absorption that would be effected by the earth's atmosphere.

The contribution of the cosmic radiation to the ionization of the atmosphere in the neighbourhood of the earth's surface, as mentioned above, is relatively small; a rough calculation shows that the essential part of the ionization of the air is accounted for here by the radioactive substances in the atmosphere (especially the α-rayers Rn, RaA, RaC, Tn, ThA, ThC). In addition, the β- and γ-radiation from the radioactive substances contained in the earth is of importance, as can be seen from the following Table 55, in which *I* denotes the formation of a pair of ions per c.c. per second.

TABLE 55

Ionization in the Neighbourhood of the Earth's Surface

By the α-, β-, and γ-rays from radio-elements in the air	4·9*I*
By the β- and γ-rays from radio-elements in the earth	3·1*I*
By the cosmic radiation	1·5*I*
Total effect of all known ionizing agents	9·5*I*

In view of the uncertainty of the bases of the calculation, the agreement with the empirical value of 10–13 ions given early in this section is satisfactory, so that it is not necessary to regard any other source of ionization, such as ultra-violet light, as of essential importance in the production of atmospheric ionization. As already mentioned, there are practically no radioactive substances present over the oceans, and accordingly it has been possible to calculate that the very slight ionization of the air over the high seas is to be regarded as primarily due to the effect of cosmic radiation.

5. The Aurora

Recent investigations have shown that the phenomena of the northern lights can be explained as a consequence of the penetration of electrons emitted by the sun into the earth's atmosphere. The occurrence of the aurora in polar regions and the obvious impossibility of their production in low latitudes, as well as the details of their form have been made intelligible as an effect of the magnetic field of the earth on the path of the electrons (see above, p. 281). However, no details are as yet known on the mechanism of the emission of electrons from the sun. Gases of the earth's atmosphere are excited to luminescence by the electrons. The existence of nitrogen is revealed spectroscopically by the presence of the first positive group of nitrogen bands in the red, and the red and green lines ($\lambda = 6,300$ and $5,577$ A.) characteristic of the aurora have been identified as oxygen lines. These results are of particular interest inasmuch as they prove that even at the greatest heights at which the northern lights have been observed in the atmosphere, i.e. up to several hundred kilometres, nitrogen and even the heavier gas oxygen are still present.

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XXVI

THE HISTORICAL DEVELOPMENT OF THE SCIENCE OF RADIOACTIVITY

On the 24th of February 1896 Henri Becquerel read a paper before the Academy of Science in Paris in which he stated that compounds of uranium emit rays that are able to affect a photographic plate through material opaque to light. This casual observation was the starting-point in the development of a new branch of knowledge, to wit, radioactivity.† After Becquerel's discovery, numerous substances were tested for similar properties to those shown by uranium, and as a result the radioactivity of thorium was discovered by C. G. Schmidt and by Mme Curie (1898). In the same year the systematic investigation of different uranium materials led Pierre Curie and his wife to the discovery of two markedly strong active substances in pitchblende, one of which was separated along with bismuth and named 'polonium', whereas the second, 'radium', was found to give similar reactions to salts of barium. Further investigation revealed the fact that it is very difficult to concentrate polonium (Marckwald), but that radium is a substance sufficiently long-lived to permit of its preparation in weighable quantities, and at the same time sufficiently short-lived to show in a high degree the typical properties of a radio-element. In addition to these radium showed several other favourable properties, such as the production of a gaseous succession product of not too short life. From amongst the most important of the remaining radioactive substances, first actinium (1899) was discovered by Debierne and by Giesel, then radio-lead by K. A. Hofmann, and radiothorium (1905) and mesothorium (1907) by Hahn. In the year 1907 ionium was discovered in uranium minerals by Boltwood and almost simultaneously by Hahn and Marckwald, and in the same year N. R. Campbell succeeded in establishing the radioactivity of potassium and rubidium. The discovery of an additional radio-element of long life, protoactinium, was not made until the year 1918 by Hahn and Meitner on one hand and by Soddy and Cranston on the other. In 1932 Hevesy and Pahl found that samarium was a radioactive substance. The production of heat by radium was observed at an early stage, but first measured in the year

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† Becquerel was investigating all kinds of fluorescent substances, because at that time a connexion was thought to exist between fluorescence and X-rays, but the new radiation was first found with fluorescent salts of uranium. He recognized later that all salts of uranium send out rays, so that fluorescence has nothing to do with the new phenomenon. As has often happened in the history of science, this discovery constituted what Lodge has termed 'deserved good fortune'.

1903 by Curie and Laborde; an especially accurate determination of this important quantity resulted later from the investigations of Schweidler, St. Meyer and Hess, and Rutherford.

By the discovery of radioactive substances a new and powerful field of natural phenomena was opened up, but at first there was lacking a satisfactory linking up of the numerous inexplicable individual phenomena with each other, as well as with the phenomena met with in other physico-chemical studies. Light was first shed on these problems by the disintegration theory, formulated by Rutherford and Soddy in the year 1902. According to this theory the emission of radiation by radioactive substances is a phenomenon consequent upon atomic transformation, and the theory made it possible to deduce quantitative information on the process of transformation from the nature and intensity of the radiation. The idea of a development of inorganic matter had frequently been conjectured in the course of the history of chemistry, and by virtue of the disintegration theory and the unbounded evidence in its favour, it has established itself permanently in the fabric of our knowledge. The disintegration theory of Rutherford and Soddy has thereby achieved the distinction of becoming one of the most important theories in natural science.

The investigations that resulted from the formulation of this theory at first proceeded mainly in two directions. First it was necessary to examine the nature of the rays from radioactive substances, and then to reveal the sequence of the transformation processes and the properties of the radio-elements.

In 1899 Giesel, St. Meyer and Schweidler, and H. Becquerel had already established the fact that part of the radioactive rays could be deflected in a magnetic field. A few years later Rutherford and his co-workers succeeded in identifying the exact nature of the α-, β-, and γ-rays. Of particular importance for the recognition of the nature of the α-rays was the work of Ramsay and Soddy, and of Rutherford and Royds, who showed that the rays are charged atoms of helium. P. Curie had already observed that they have a definite range, and the study of their properties was mainly the work of W. H. Bragg, Geiger, Marsden, Kleeman, Mme Curie and her collaborators, Rausch von Traubenberg, and others. Elster and Geitel on one hand, and Crookes on the other, made the observation that α-rays are able to excite scintillations in zinc sulphide screens, and this proved to be of great practical importance. Regener was the first to make quantitative counts of scintillations, and in a similar way Rutherford and Geiger succeeded in determining the number of α-particles emitted per second by 1 gm. of radium, and also the charge of an α-particle. Schweidler was the first to direct attention to the statistical nature of radioactive disintegration.

The more detailed elucidation of the properties of β-rays was carried out in the laboratories of Lenard, J. J. Thomson, Rutherford, Hahn, Meitner, v. Baeyer, Geiger, Bothe, Danysz, de Broglie, and Kovarik; that of γ-rays by Rutherford and his collaborators, Ellis in particular, by W. H. Bragg, A. H. Compton, Meitner, and as a result of investigations in the Vienna Radium Institute (K. W. F. Kohlrausch, Hess, Lawson, and others). The method of C. T. R. Wilson for rendering visible the tracks of the rays by condensation of water vapour has been of the greatest service in the study of all three types of rays.

The phenomenon of recoil, which was discovered by Hahn and by Russ and Makower and later investigated particularly by Wertenstein, made possible the isolation of several elements of short life. The relation between range and disintegration constant, discovered by Geiger and Nuttall in 1911, has proved very important in revealing the existence and for determining the period of elements both of very short and of very long life.

The nuclear theory of the atom, which is of paramount importance in modern physics, was advanced in the same year by Rutherford. It was based on observations by Geiger and Marsden on the scattering of α-rays in their passage through matter, and furnished a satisfactory explanation of this phenomenon. But on the basis of the classical ideas of theoretical physics it was not possible to interpret the properties or the stability of such a structure. Not until two years later was this achieved by Bohr, who utilized Planck's quantum theory for this purpose. The atomic theory founded by Bohr made it possible for the first time to give a simple and numerical interpretation of absorption and emission spectra, to which phase of progress the work of Sommerfeld and his school, Kramers, and others belongs, and also elucidated the extensive field of the phenomena of collisions between electrons and atoms, which was initiated by the work of Franck and Hertz. The further development of the theory of atomic structure was naturally linked most intimately with the fundamental successes of quantum mechanics; for this reason mention must be made primarily of the names of Born, L. de Broglie, Dirac, Heisenberg, Pauli, and Schrödinger.

The development of the study of X-rays is also closely related to the modern theory of the atom. The famous discovery of v. Laue (1912) was soon followed by the work of W. H. and W. L. Bragg on the reflection of X-rays by crystals, and by Moseley's investigation on the characteristic X-ray lines of the chemical elements, by virtue of which the fundamental importance of the atomic number was established (1913). But radioactivity itself also owes a debt to the development of X-ray spectroscopy. Thus the measurement of the wave-lengths of γ-rays, as carried out by Ellis and by Meitner, first became a practicable proposition when the spectroscopy of X-rays had made it possible to localize the individual groups of electrons in the extra-nuclear part of the atom, and to calculate the work necessary to separate an electron from the atom. Only in the case of soft γ-rays had Rutherford and Andrade succeeded previously in carrying out a determination of the frequency by the crystal method.

Meanwhile, important advances had also been made in radiochemistry; investigations by Marckwald, Keetmann, Boltwood, Auer v. Welsbach, McCoy, Strömholm and Svedberg, Fleck, and others led to a conception held notably by Soddy as early as 1910, namely, the existence of radioactive elements, later called 'isotopes', which, in spite of differences in their atomic weights and radioactive properties, at the same time show the same chemical, electrochemical, and spectroscopic properties. On the basis of this view, and with the aid of more complete knowledge of the sequence of disintegration in the transformation series of the radio-elements, the radioactive displacement laws were formulated by A. S. Russell, and almost simultaneously by Fajans and by Soddy (1913), the results of the last two authors being satisfactory in every detail. Amongst other things, these laws made it possible to settle the question of the end-products of the disintegration series, and the conclusions formed in the course of this work were completely confirmed by atomic-weight determinations carried out by Richards, Hönigschmid, and others. It became more and more clear that these results are of the greatest importance not only in radiochemistry, but also in general chemistry. The conception of a chemical element had to be restricted to its chemical inseparability, and even in the case of the ordinary elements it became imperative to reckon with the possibility that they might consist of more than one type of atom.

A few years later Aston proved by means of his mass-spectrograph that the phenomenon of isotopy is actually a general property of the elements. This apparatus was constructed by Aston as a further development of the pioneer work of J. J. Thomson on positive-ray analysis. At the same time Aston's investigations (1919) revealed the fact that, to a first approximation, the atomic weights of the elements are integral in nature, even when their combining weights often show marked deviations from whole-number values. This invalidated the most important argument that perforce had always been raised against Prout's hypothesis, and rendered it probable that all the chemical elements are genetically related. Science was confronted with a new problem, that of separating the elements which occur in nature into their constituent isotopes, and in several cases this problem has been more or less completely solved. Owing to its singular properties, the heavy isotope of hydrogen was prepared in the pure state soon after its discovery by Urey. For other elements, the best results have been attained by G. Hertz in Charlottenburg, by use of his diffusion method.

The first successful experiments to effect a transmutation of the non-radioactive elements also fall into the post-war period. Success was first achieved in the case of nitrogen in 1919, when Rutherford and Chadwick demonstrated that by bombardment with α-particles hydrogen nuclei (protons) are liberated from the atoms of this element, these protons being projected as corpuscular rays of large range. Further investigations have established that a large number of light elements can be transformed artificially in this way. In addition to the above-mentioned discoverers of artificial disintegration, many other workers have taken part in the subsequent development of the subject, including Kirsch, Pettersson, and Stetter in Vienna, Harkins in Chicago, Bothe in Berlin, and Pose in Halle. These studies have led to important conclusions on the structure of atomic nuclei, the constitution of which had to remain a closed book at the time of the formulation of the theory of the atom due to Rutherford and Bohr. They have not only introduced us to the proton as a nuclear constituent, but during the course of the last few years also to two further constituents of matter, the neutron and the positron. The neutron was discovered in 1932 by Chadwick by bombarding beryllium with α-rays, as a sequel to observations of Bothe and of M. and Mme Curie-Joliot. The positron was discovered in the same year by Anderson, and it was observed with greater certainty in 1933 by Blackett and Occhialini, using a Wilson cloud chamber, as a product of the interaction of cosmic rays on matter.

Although for many years the only available means of transmuting atomic nuclei were the α-rays from radioactive substances, Cockroft and Walton succeeded in 1932 in the transmutation of light elements, by bombarding them with a beam of fast protons that had been accelerated in a discharge tube. This was the first time that a transmutation of elements had been achieved without the intervention of a natural radioactive process. The methods of accelerating the protons were varied in many ways, and in the year 1932 an ingenious device called a cyclotron (see p. 290) was developed by E. O. Lawrence in California. This was also the first method to show that deuterons of mass 2 are more efficient projectiles than protons of mass 1.

The experiments on the transmutation of elements entered a particularly interesting phase when, in 1933, M. and Mme Curie-Joliot found that the product of atomic transformation in certain cases is radioactive. The development of this fundamental discovery resulted in the establishment, within a year or two, of a new branch of science, that of artificial radioactivity. In this rapid and brilliant development the two most outstanding events were those associated with the names of Fermi and of Lawrence. Fermi found that artificially radioactive substances can be produced from most elements in the periodic system by bombarding them with neutrons, whereas Lawrence, by making use of his cyclotron (p. 290), succeeded in preparing artificial radioactive preparations of very great strength. As a result of these advances it also became possible to extend the periodic system of the elements by preparing radioactive isotopes of elements with an atomic number higher than that of uranium. Such 'trans-uranium' elements were first prepared by Fermi, and later studied in detail and with great success by Hahn and Meitner.

Successes have to be recorded in various other fields in radioactivity. The chemical effect of the rays from radium was studied in greatest detail by Lind and his co-workers, but also by Duane and Wendt, Kailan, Przibram, Mund, and others. Since 1913 radioactive indicators have frequently been utilized by Hevesy and Paneth in chemical and physical investigations; as a result of the discovery of artificial radioelements, the number of problems whose study can be simplified by the use of radioactive indicators has increased enormously, especially in the field of biology. The applications of radioactivity to geological problems have been primarily the work of Boltwood, Strutt (Lord Rayleigh), Mügge, Joly, Holmes, Urry, and Ellen Gleditsch; whilst in the domain of atmospheric electricity mention should be made of the work of Elster and Geitel, v. Schweidler, Mache, Eve, McLennan, C. T. R. Wilson, Gockel, and Hess. With the name of Hess is also associated the particularly interesting discovery of an extra-terrestrial radiation of exceeding penetrability, which he detected in balloon flights. In addition to Hess, Kolhörster, Millikan, Hoffmann, Regener, Steinke, A. H. Compton, Blackett, and Clay, in particular, have taken part in the further study of this 'cosmic radiation', and the results of their observations are no less important in astrophysics than in atomic research. In these investigations, as well as in many other recent radioactive measurements, the electronic counter due to Geiger has played a prominent part. The suitability of the photographic plate for the direct study of ray tracks has been established, notably by the Misses Blau and Wambacher (Vienna).

This short historical survey may have served to show that, in the span of only 42 years since Becquerel's discovery, the science of radioactivity has developed from a special field of physics into one of the most important and most richly correlated branches of the entire realm of natural science.

APPENDIX

THE CYCLOTRON. (*See Index, p. 297*)

SINCE the achievement of nuclear disruption and of artificial radioactivity, it has been the aim of physicists to secure bombarding ions of ever greater energy, in order to facilitate the production of the nuclear processes involved. The straightforward method of doing this by the use of larger and larger accelerating potentials for the ions, however, is fraught with progressively increasing experimental difficulties. Moreover, at these very high potentials of the order of a million volts, it is no easy task to produce concentrated beams of the ions in quantities adequate for many of the problems involved. These difficulties have been evaded in the magnetic resonance accelerator or *cyclotron* by the development of the multiple acceleration of ions to high speeds without the use of abnormally high voltages.

In this method, which was initiated and developed largely by the work of E. O. Lawrence and M. S. Livingston, semicircular hollow plates or dees, resembling the duants or binants of an electrometer, are mounted in a vacuum with their diametral edges adjacent, and are situated in a uniform magnetic field normal to the plates. High frequency oscillations are applied to the plate electrodes, and produce an oscillating electric field over the uniform diametral gap between them. Ions, formed in the middle of the diametral region by accelerated electrons from a heated spiral filament, are accelerated during one half-cycle of the oscillating electric field into the interior of one of the electrodes, where they are bent into circular paths by the magnetic field and later emerge again into the gap between the electrodes. By adjustment of the magnetic field so that the time required for the ions to traverse a semicircular path within one of the electrodes is equal to a half-period of the oscillations, the electric field will have reversed its direction when the ions return to the region between the electrodes, so that they receive a second increment of velocity on passing to the other electrode. The time required for the ions to traverse a semicircular path is independent both of the radius of the path and of the velocity of the ions, and hence, when the magnetic field has been correctly adjusted, the ions spiral round in resonance with the oscillating field until they reach the periphery of the apparatus. Their final kinetic energies are as many times greater than that corresponding to the voltage applied to the electrodes as the number of times they have crossed from one electrode to the other. Both the magnetic and electric fields exercise a focusing action on the ions and prevent any serious loss of ions as they are accelerated, so that relatively good yields of high speed ion currents are obtainable in this indirect manner, and the focusing action results in the generation of beams of ions *c.* 1 to 3 mm. in diameter, which are ideal for experimental studies of collision processes. By means of a suitably arranged electric field the high speed ions can be deflected from the accelerating system and withdrawn completely from the apparatus into the air through a thin platinum window in the chamber wall.

In one of the earlier forms of the apparatus the pole faces of the magnet were 11 inches in diameter, and a current of 10^{-9} ampere of $1{\cdot}22 \times 10^6$ e.V. protons was produced in a tube to which the maximum applied voltage was only 4,000 volts. The high frequency voltage used in a more recent form of the apparatus is estimated to be from 50,000 to 100,000 volts, and with magnet pole faces 27½ inches in diameter the ions are withdrawn from the accelerating electrodes at a distance of *c.* 12 inches from the centre of the system. With this apparatus pencils of many micro-amperes of deuterons having energies up to $6{\cdot}3 \times 10^6$ e.V. and of a few tenths of a micro-ampere of 11×10^6 e.V. doubly charged helium ions have been obtained. With such amounts of high speed deuterons incident on a beryllium target, the usual neutron emission with this apparatus is equivalent to the γ-radiation from about 100 gm. of radium, and for the protection of the operator (see pp. 258 and 260) the cyclotron is controlled at a distance of 40 feet from the target with suitable intervening absorbing material. Bombardment of sodium metal for one day with 20 micro-amperes of 5×10^6 e.V. deuterons produces radio-sodium having a γ-ray activity equivalent to that of 200 mg. of radium. In some of the recent forms of the apparatus the strength of the magnetic field is of the order of 18,000 oersteds, and the corresponding wave-length of the oscillating electric field used for accelerating deuterons or doubly charged helium ions is about 22 metres.

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TABLE 56

Artificially produced Radioactive Isotopes of the Elements†

Element and atomic number	*Radioactive isotopes*		
	Mass-numbers	*Half-value periods*†	*Particles emitted*†
2 He	6	1 s.	(−)
3 Li	8	0·8 s.	(−)
4 Be	10	> 10 y.	(−)
5 B	12	0·02 s.	(−)
6 C	11	20 m.	(+)
7 N	13; 16	10·5 m.; 9 s.	(+); (−)
8 O	15; 19	2·1 m.; 31 s.	(+); (−)
9 F	17; 18; 20	1·2 m.; 108 m.; 8 s.	(+); (+); (−)
10 Ne	23	33 s.	(−)
11 Na	22; 24	3 y.; 15 h.	(+); (−)
12 Mg	27	12 m.	(−)
13 Al	26; 28; 29	7 s.; 2·3 m.; 11 m.	(+); (−); (−)
14 Si	27; 31	6·6 m.; 2·5 h.	(+); (−)
15 P	30; 32	3 m.; 14·5 d.	(+); (−)
16 S	31; 35	26 m.; 80 d.	(+); (−)
17 Cl	34; 38	33 m.; 38 m.	(+); (−)
18 A	41	110 m.	(−)
19 K	38; 42	7·5 m.; 12·5 h.	(+); (−)
20 Ca	39; 45	4·5 m.; 2·3 h.	(+); (−)
21 Sc	41; 42; 43; 44; 46	53 m.; 4·1 h.; 4·0 h.; 52 h.;	(+); (+); (+); (+); (−);
	48	90 d.; 41 h.	(−)
22 Ti	51	3 m.	(−)
23 V	48; 49; 50; 52	16 d.; 32 m.; 3·6 h.; 3·8 m.	(+); (+); (+); (−)
24 Cr			
25 Mn	?; 56; ?; ?; ?	46 m.; 2·5 h.; 21 m.; 5 d.; sev. mths.	(+); (−); (−); (−); (−)
26 Fe	55; 59	8·9 m.; 40 d.	(+); (−)
27 Co	55; ?; 58; 60	18 h.; 150 d.; 11 m.; c. 1 y.	(+); (+ and −); (+); (−)
28 Ni	63	160 m.	(−)
29 Cu	61; 62; 64; 66	3·3 h.; 10·5 m.; 12·5 h.; 5 m.	(+); (+); (+ and −); (−)
30 Zn	63; 65	38 m.; 60 m.	(+); (−)
31 Ga	66; 68; 70; 72	9·4 h.; 60 m.; 20 m.; 23 h.	(+); (+); (−); (−)
32 Ge	75; ?	20 h.; 30 m.	(−); (−)
33 As	76; 78	26 h.; 65 m.	(−); (−)
34 Se	79 or 81; 83	1 h.; 17 m.	(−); (−)
35 Br	78; 80; 80; 82; 83	6·3 m.; 18 m.; 4·5 h.; 34 h.; 2·5 h.	(+); (−); (−); (−); (−)
36 Kr	?	74 m.; 4·5 h.; 18 h.	(−); (−); (?)
37 Rb	86; 88	18 m.; 18 d.	(−); (−)
38 Sr	89	3 h.; 55 d.	(−); (−)
39 Y	90	70 h.	(−)
40 Zr	97	44 h.	(−)
41 Nb			
42 Mo	?	2·5 m.; 17 m.; 36 h.	

TABLE 56 (*continued*)

Element and atomic number	*Radioactive isotopes*		
	Mass-numbers	*Half-value periods*†	*Particles emitted*†
43 —	?	few months	
44 Ru	?	40 s.; 100 s.; 11 h.; 170 h.	(−); (−); (−); (−)
45 Rh	?	44 s.; 3·9 m.	
46 Pd	?	60 h.; 15 m.; 12 h.; 3 m.	
47 Ag	106; 106; 108; 110; 111; 112	24·5 m.; 8·2 d.; 2·3 m.; 22 s.; 7·5 d.; 3·2 h.	(+); (−); (−); (−); (−); (−)
48 Cd	?; 115; 117	33 m.; 4·3 h.; 58 h.	(+); (−); (−)
49 In	111; 112; 114; 114 116; 116; 117	20 m.; 72 s.; 4·1 h.; 50 d. 13 s.; 54 m.; 2·3 h.	(+); (−); (−); (−); (−); (−); (−)
50 Sn	?	8 m.; 18 m.	
51 Sb	120; 122; 124	16 m.; 2·5 d.; 60 d.	(+); (−); (−)
52 Te	?	1·1 h.	(−)
53 I	128	25 m.	(−)
54 X			
55 Cs	134	1·5 h.	(−)
56 Ba	?; 139	2·5 m.; 80 m.	(?); (?)
57 La	140	1·3 d.	(−)
58 Ce	139	2·1 m.	(+)
59 Pr	140; 142	3 m.; 19 h.	(+); (−)
60 Nd	147; 149; 151	84 h.; 2 h.; 21 m.	(−); (−); (−)
61 —			
62 Sm	151; 153	21 m.; 2 d.	(−); (−)
63 Eu	152; 154	27 h; 9·3 h.	(+); (−)
64 Gd	159	3·5 m; 17 h.	(−); (−)
65 Tb	160	3·9 h.	(−)
66 Dy	159; 165	2·2 m.; 2·5 h.	(+); (−)
67 Ho	164; 166	47 m.; 30 h.	(+); (−)
68 Er	165; 169; 171	5 h.; 7 m.; 12 h.	(+); (−); (−)
69 Tm	170	120 d.	(−)
70 Yb	175; 175	2·1 h.; 41 h.	(−); (−)
71 Lu	176; 178	4 h.; 8 d.	(−); (−)
72 Hf	181	55 d.	(−)
73 Ta	180; 182	8 h.; 97 d.	(+); (−)
74 W	?	1 d.	
75 Re	?; 188; ?	20 m.; 18 h.; 85 h.	(?); (−); (−)
76 Os	?	40 h.	
77 Ir	192; 192 or 194; 189, 192, or 195	2 m.; 1·5 m. and 19 h.; 28 m. and 8·5 h.	(?); (−); (−); (−); (−)
78 Pt	197; 199	18 h. and 3·3 d.; 50 m.	(−); (−)
79 Au	198; 199	2·7 d.; 13 h. and 4·5 d.	(−); (−); (−)
80 Hg	205; 206	41 h.; 45 m.	(−); (−)
81 Tl	204; 206	5 m.; 97 m.	(−); (−)
82 Pb	209	3 h.	(−)
83 Bi			
. . . . .			
. . . . .			
. . . . .			
. . . . .			
88 Ra	229	1 m.	(−)
89 Ac	231; 232; 233	12 m.; 42 h.; 3·5 h.	(−); (?); (−)
90 Th	239 (233 ?)	28 m.	(−)
91 Pa	233	2·5 m.	(−)

TABLE 56 (*continued*)

Element and atomic number	*Radioactive isotopes*		
	Mass-numbers	*Half-value periods*†	*Particles emitted*†
92 U	236 ?; 239	23 m.; 10 s. and 40 s.	(−); (−)
93 Eka Re	239	2·2 m. and 16 m.	(−)
94 Eka Os	239	59 m.; 17 h. and 5·7 h.	(−); (−)
95 Eka Ir	239	66 h.	(−)
96 Eka Pt	239	2·5 h.	(−)

† In connexion with this table, reference should be made to the mass-numbers and abundance of the stable isotopes given in Table 43, p. 182, as well as to the text on p. 120, and to Fig. 40 (p. 121). See also Table 29, p. 116, which is restricted to radioactive isotopes produced by *neutron* bombardment. (−) = electron (β-ray); (+) = positron; s. = second; m. = minute; h. = hour; d. = day; y. = year.

TABLE 57

Constants frequently used in Radioactive Calculations

Velocity of Light $c = 2{\cdot}9978 \times 10^{10}$ cm./sec.
Planck's Constant $h = 6{\cdot}610 \times 10^{-27}$ erg sec.
Charge of Electron $e = 4{\cdot}800 \times 10^{-10}$ e.s.u. $= 1{\cdot}6012 \times 10^{-20}$ e.m.u.
Mass of Electron $m = 9{\cdot}105 \times 10^{-28}$ gm.
Mass of Hydrogen Atom $m_H = 1{\cdot}673 \times 10^{-24}$ gm.

Unit of Atomic Weight $m_1 = \dfrac{m_H}{1{\cdot}0081} = 1{\cdot}660 \times 10^{-24}$ gm.

$m_1 = 0{\cdot}93105 \times 10^9$ e.V.; 1 e.V. $= 1{\cdot}0741 \times 10^{-9}\, m_1$.

Atomic Weight in Chemical Scale = 0·99973 × Atomic Weight in Physical Scale.
Atomic Weight in Physical Scale = 1·00027 × Atomic Weight in Chemical Scale.
Gram-Molecular Volume (at N.T.P.) $v_0 = 2{\cdot}2414 \times 10^4$ c.c.
Loschmidt's Number (Avogadro's Number):

per mol $N = 6{\cdot}028 \times 10^{23}$;
per c.c. (at N.T.P.) $L = 2{\cdot}689 \times 10^{19}$ cm.$^{-3}$

Mass of 1 c.c. Hydrogen (at N.T.P.) $= 8{\cdot}987 \times 10^{-5}$ gm.;
Volume of 1 gm. Hydrogen (at N.T.P.) $= 1{\cdot}1127 \times 10^4$ c.c.
Mass of 1 c.c. Helium (at N.T.P.) $= 1{\cdot}785 \times 10^{-4}$ gm.;
Volume of 1 gm. Helium (at N.T.P.) $= 5{\cdot}6023 \times 10^3$ c.c.

1 Year $= 3{\cdot}65 \times 10^2$ days $= 8{\cdot}760 \times 10^3$ hours $= 5{\cdot}256 \times 10^5$ minutes $= 3{\cdot}1536 \times 10^7$ seconds.

1 Day $= 1{\cdot}44 \times 10^3$ minutes $= 8{\cdot}64 \times 10^4$ seconds.

$$e = 2{\cdot}71828; \quad \frac{1}{e} = 0{\cdot}36788; \quad \log_{10} e = 0{\cdot}43429; \quad \log_{10} x = 0{\cdot}43429 \log_e x.$$

$$\log_e 10 = \frac{1}{\log_{10} e} = 2{\cdot}30259; \quad \log_e x = 2{\cdot}30259 \log_{10} x.$$

$$\log_e 2 = 0{\cdot}69315; \quad \frac{1}{\log_e 2} = 1{\cdot}44270.$$

For Atomic Weights of the Elements see Table 43 (p. 182).

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