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Marie Curie Nobel Lecture

Nobel Lecture, December 11, 1911*

Radium and the New Concepts in Chemistry

Some 15 years ago the radiation of uranium was discovered by Henri Becquerel¹, and two years later the study of this phenomenon was extended to other substances, first by me, and then by Pierre Curie and myself². This study rapidly led us to the discovery of new elements, the radiation of which, while being analogous with that of uranium, was far more intense. All the elements emitting such radiation I have termed *radioactive*, and the new property of matter revealed in this emission has thus received the name *radioactivity*. Thanks to this discovery of new, very powerful radioactive substances, particularly radium, the study of radioactivity progressed with marvellous rapidity: Discoveries followed each other in rapid succession, and it was obvious that a new science was in course of development. The [Swedish Academy of Sciences](#) was kind enough to celebrate the birth of this science by awarding the Nobel Prize for Physics to the first workers in the field, [Henri Becquerel, Pierre Curie and Marie Curie](#) (1903).

judgement, and the boldness of his hypotheses and through the many investigations carried out by him and his pupils, has succeeded not only in increasing our knowledge but also in classifying it with great clarity; he has provided a backbone for the new science, in the form of a very precise theory admirably suited to the study of the phenomena. I am happy to recall that [Rutherford](#) came to Stockholm in 1908 to receive the Nobel Prize as a well-deserved reward for his work.

Far from halting, the development of the new science has constantly continued to follow an upward course. And now, only 15 years after Becquerel's discovery, we are face to face with a whole world of new phenomena belonging to a field which, despite its close connexion with the fields of physics and chemistry, is particularly well-defined. In this field the importance of radium from, the viewpoint of general theories has been decisive. The history of the discovery and the isolation of this substance has furnished proof of my hypothesis *that radioactivity is an atomic property of matter and can provide a means of seeking new elements*. This hypothesis has led to present-day theories of radioactivity, according to which we can predict with certainty the existence of about 30 new elements which we cannot generally either isolate or characterize by chemical methods. We also assume that these elements undergo atomic transformations, and the most direct proof in favour of this theory is provided by the experimental fact of the formation of the chemically defined element *helium* starting from the chemically-defined element *radium*.

Viewing the subject from this angle, it can be said that the task of isolating radium is the corner-stone of the edifice of the science of radioactivity. Moreover, radium remains the most useful and powerful tool in radioactivity laboratories. I believe that it is because of these considerations that the Swedish Academy of Sciences has done me the very great honour of awarding me this year's Nobel Prize for Chemistry.

It is therefore my task to present to you radium in particular as a new chemical element, and to leave aside the description of the many radioactive phenomena which have already been described in the Nobel Lectures of H. Becquerel, P. Curie and E. Rutherford.

Before broaching the subject of this lecture, I should like to recall that the discoveries of radium and of polonium were made by Pierre Curie in collaboration with me. We are also indebted to Pierre Curie for basic research in the field of radioactivity, which has been carried out either alone, in collaboration with his pupils.

connected with our common work. I thus feel that I interpret correctly the intention of the Academy of Sciences in assuming that the award of this high distinction to me is motivated by this common work and thus pays homage to the memory of Pierre Curie.

I will remind you at the outset that one of the most important properties of the radioactive elements is that of ionizing the air in their vicinity (Becquerel). When a uranium compound is placed on a metal plate A situated opposite another plate B and a difference in potential is maintained between the plates A and B, an electric current is set up between these plates; this current can be measured with accuracy under suitable conditions and will serve as a measure of the activity of the substance. The conductivity imparted to the air can be ascribed to ionization produced by the rays emitted by the uranium compounds.

In 1897, using this method of measurement, I undertook a study of the radiation of uranium compounds, and soon extended this study to other substances, with the aim of finding out whether radiation of this type occurs in other elements. I found in this way that of the other elements known, only the compounds of thorium behave like the compounds of uranium.

I was struck by the fact that the activity of uranium and thorium compounds appears to be *an atomic property of the element uranium and of the element thorium*. Chemical compounds and mixtures containing uranium and thorium are active in direct proportion to the amount of these metals contained in them. The activity is not destroyed by either physical changes of state or chemical transformations.

I measured the activity of a number of minerals; all of them that appear to be radioactive always contain uranium or thorium. But an unexpected fact was noted: certain minerals (pitchblende, chalcocite, autunite) had a greater activity than might be expected on the basis of their uranium or thorium content. Thus, certain pitchblendes containing 75% of uranium oxide are about four times as radioactive as this oxide. Chalcocite (crystallized phosphate of copper and uranium) is about twice as radioactive as uranium. This conflicted with views which held that no mineral should be more radioactive than metallic uranium. To explain this point I prepared synthetic chalcocite from pure products, and obtained crystals, whose activity was completely consistent with their uranium content; this activity is about half that of uranium.

thorium and the elements known at present. It also occurred to me that if this was the case I might be able to extract this substance from the mineral by the ordinary methods of chemical analysis. Pierre Curie and I at once carried out this research, hoping that the proportion of the new element might reach several per cent. In reality the proportion of the hypothetical element was far lower and it took several years to show unequivocally that pitchblende contains at least one highly-radioactive material which is a new element in the sense that chemistry attaches to the term.

We were thus led to create a new method of searching for new elements, *a method based on radioactivity considered as an atomic property of matter*. Each chemical separation is followed by a measurement of the activity of the products obtained, and in this way it is possible to determine how the active substance behaves from the chemical viewpoint. This method has come into general application, and is similar in some ways to spectral analysis. Because of the wide variety of radiation emitted, the method could be perfected and extended, so that it makes it possible, not only to discover radioactive materials, but also to distinguish them from each other with certainty.

It was also found in using the method being considered, that it was in fact possible to concentrate the activity by chemical methods. We found that pitchblende contains at least two radioactive materials, one of which, accompanying bismuth, has been given the name *polonium*, while the other, paired with barium, has been called *radium*.

Other radioactive elements have been discovered since: actinium (Debiere), radiothorium and mesothorium (Hahn), ionium (Boltwood), etc.

We were convinced that the materials which we had discovered were new chemical elements. This conviction was based solely on the atomic nature of radioactivity. But at first, from the chemical viewpoint, it was as if our substances had been, the one pure bismuth, and the other pure barium. It was vital to show that the radioactive property was connected with traces of elements that were neither bismuth nor barium. To do that the hypothetical elements had to be isolated. In the case of radium isolation was completely successful but required several years of unremitting effort. Radium in the pure salt form is a substance the manufacture of which has now been industrialized; for no other new radioactive substance have such positive results been obtained.

The radiferous minerals are being subjected to very keen study because the presence of radium lends them considerable value. They are identifiable either by the electrometric

long time been processed to yield uranium salts. After extraction of the latter, the mineral leaves a residue which contains radium and polonium. We have normally used this residue as our raw material.

The first treatment consists in extracting the radiferous barium and the bismuth containing the polonium. This treatment, which was first performed in the laboratory on several kilograms of raw material (as many as 20 kg) had then to be undertaken in a factory owing to the need to process thousands of kilograms. Actually, we gradually learned from experience that the radium is contained in the raw material in the proportion of a few decigrams per ton. About 10 to 20 kg crude barium sulphate containing radium are extracted from one ton of residue. The activity of these sulphates is even then 30 to 60 times greater than that of uranium. These sulphates are purified and converted to chlorides. In the mixture of barium and radium chlorides the radium is present only in the proportion of about 3 parts per 100,000. In the radium industry in France a much lower grade mineral is most often used and the proportion indicated is far lower still. To separate the radium from the barium I have used a method of fractional crystallization of the chloride (the bromide can also be used). The radium salt, less soluble than the barium salt, becomes concentrated in the crystals. Fractionation is a lengthy, methodical operation which gradually eliminates the barium. To obtain a very pure salt I have had to perform several thousands of crystallizations. The progress of the fractionation is monitored by activity measurements.

A first proof that the element radium existed was furnished by spectral analysis. The spectrum of a chloride enriched by crystallization exhibited a new line which Demarcay attributed to the new element. As the activity became more concentrated, the new line increased in intensity and other lines appeared while the barium spectrum became at the same time less pronounced. When the purity is very high the barium spectrum is scarcely visible.

I have repeatedly determined the average atomic weight of the metal in the salt subjected to spectral analysis. The method used was the one consisting in determining the chlorine content in the form of silver chloride in a known amount of the anhydrous chloride. I have found that this method gives very good results even with quite small amounts of substance (0.1 to 0.5 g), provided a very fast balance is used to avoid the absorption of water by the alkaline-earth salt during the weighings. The atomic weight increases with the enrichment of the radium as indicated by the spectrum. The successive atomic weights obtained were: 138; 146; 174; 225; 226.45. This last value was

experiments.

The preparation of pure radium salts and the determination of the atomic weight of radium have proved positively that radium is a new element and have enabled a definite position to be assigned to it. Radium is the higher homologue of barium in the family of alkaline-earth metals; it has been entered in Mendeleev's table in the corresponding column, on the row containing uranium and thorium. The radium spectrum is very precisely known. These very clear-cut results for radium have convinced chemists and justified the establishment of the new science of radioactive substances.

In chemical terms radium differs little from barium; the salts of these two elements are isomorphic, while those of radium are usually less soluble than the barium salts. It is very interesting to note that strong radioactivity of radium involves no chemical anomalies and that the chemical properties are actually those which correspond to the position in the Periodic System indicated by its atomic weight. The radioactivity of radium in solid salts is ca. 5 million times greater than that of an equal weight of uranium. Owing to this activity its salts are spontaneously luminous. I also wish to recall that radium gives rise to a continuous liberation of energy which can be measured as heat, being about 118 calories per gram of radium per hour.

Radium has been isolated in the metallic state (M. Curie and A. Debierne, 1910). The method used consisted in distilling under very pure hydrogen the amalgam of radium formed by the electrolysis of a chloride solution using a mercury cathode. One decigram only of salt was treated and consequently considerable difficulties were involved. The metal obtained melts at about 700°C, above which temperature it starts to volatilize. Is it very unstable in the air and decomposes water vigorously.

The radioactive properties of the metal are exactly the ones that can be forecast on the assumption that the radioactivity of the salts is an atomic property of the radium which is unaffected by the state of combination. It was of real importance to corroborate this point as misgivings had been voiced by those to whom the atomic hypothesis of radioactivity was still not evident.

Although radium has so far only been obtained in very small amounts, it is nevertheless true to say, in conclusion, that it is a perfectly defined and already well-studied chemical element.

proportion of polonium in the mineral is about 5,000 times smaller than that of radium.

Before theoretical evidence was available from which to forecast this proportion, I had conducted several extremely laborious operations to concentrate polonium and in this way had secured products with very high activity without being able to arrive at definite results as in the case of radium. The difficulty is heightened by the fact that polonium disintegrates spontaneously, disappearing by half in a period of 140 days. We now know that radium has not an infinite life either, but the rate of disappearance is far less (it disappears by half in 2,000 years). With our facilities we can scarcely hope to determine the atomic weight of polonium because theory foresees that a rich mineral can contain only a few hundredths of a milligram per ton, but we can hope to observe its spectrum. The operation of concentrating polonium, as I shall point out later, is, moreover, a problem of great theoretical interest.

Recently, in collaboration with Debierne, I undertook to treat several tons of residues from uranium mineral with a view to preparing polonium. Initially conducted in the factory, then in the laboratory, this treatment finally yielded a few milligrams of substance about 50 times more active than an equal weight of pure radium. In the spectrum of the substance some new lines could be observed which appear attributable to polonium and of which the most important has the wavelength 4170.5 \AA . According to the atomic hypothesis of radioactivity, the polonium spectrum should disappear at the same time as the activity and this fact can be confirmed experimentally,

I have so far considered radium and polonium only as chemical substances. I have shown how the fundamental hypothesis which states that radioactivity is an atomic property of the substance has led to the discovery of new chemical elements. I shall now describe how the scope of this hypothesis has been greatly enlarged by the considerations and experimental facts which resulted in establishing the theory of atomic radioactive transformations.

The starting-point of this theory must be sought in the considerations of the source of the energy involved in the phenomena of radioactivity. This energy becomes manifest as an emission of rays which produce thermal, electrical and light phenomena. As the emission occurs spontaneously without any known cause of excitation, various hypotheses have been advanced to account for the liberation of energy. One of the hypotheses put forward at the beginning of our research by Pierre Curie and myself consisted in assuming that the radiation is an emission of matter accompanied by a loss

This hypothesis, which at first could only be enunciated together with other equally valid theories, has attained dominant importance and finally asserted itself in our minds owing to a body of experimental evidence which substantiated it. This evidence is essentially the following: A series of radioactive phenomena exists in which radioactivity appears to be tied up to matter in an imponderable quantity, the radiation moreover not being permanent but disappearing more or less rapidly with time. Such are polonium, radioactive emanations and deposits of induced radioactivity.

It has been established moreover in certain cases that the radioactivity observed increases with time. This is what happens in the case of freshly prepared radium, of the emanation freshly introduced into the measuring apparatus, of thorium deprived of thorium X, etc.

A careful study of these phenomena has shown that a very satisfactory general explanation can be given by assuming that each time a decrease of radioactivity is observed there is a destruction of radioactive matter, and that each time an increase of activity is observed, there is a production of radioactive matter. The radiations which disappear and appear are, besides, of very varied nature and it is admitted that every kind of rays determined can serve to characterize a substance which is its source, and appears and disappears with it.

As radioactivity is in addition a property which is essentially atomic, the production or the destruction of a distinct type of radioactivity corresponds to a production or a destruction of atoms of a radioactive substance.

Finally, if it is supposed that radioactive energy is a phenomenon which is borrowed from atomic transformation, it can be deduced from this that every radioactive substance undergoes such a transformation, even though it appears to us to be invariable. Transformation in this case is only very slow and this is what takes place in the case of radium or uranium.

The theory I have just summarized is the work of Rutherford and [Soddy](#), which they have called *theory of atomic disintegration*. By applying this theory it can be concluded that a primary radioactive substance such as radium undergoes a series of atomic transmutations by virtue of which the atom of radium gives birth to a train of atoms of smaller and smaller weights, since a stable state cannot be attained as long as the atom formed is radioactive. Stability can only be attained by inactive matter.

end-products of the evolution of radium, and that it is in the form of alpha rays that the helium atoms which are formed when radium atoms disintegrate are discharged. Now, the production of helium by radium has been proved by the experiments of [Ramsay](#) and Soddy, and it cannot now be contested that the perfectly defined chemical element, radium, gives rise to the formation of another equally defined element – helium. Moreover, the investigations done by Rutherford and his students have proved that the alpha particles emitted by radium with an electric charge are also to be found in the form of helium gas in the space where they have been recovered.

I must remark here that the bold interpretation of the relationship existing between radium and helium rests entirely upon the certitude that radium has the same claim to be a chemical element as have all the other known elements, and that there can be no question of regarding it to be a molecular combination of helium with another element. This shows how fundamental in these circumstances has been the work carried out to prove the chemical individuality of radium, and it can also be seen in what way the hypothesis of the atomic nature of radioactivity and the theory of radioactive transformations have led to the experimental discovery of a first clearly-established example of atomic transmutation. This is a fact the significance of which cannot escape anyone, and one which incontestably marks an epoch from the point of view of chemists.

Considerable work, guided by the theory of radioactive transformations, has led to approximately 30 new radioactive elements being envisaged, classified in 4 series according to the primary substance: these series are uranium, radium, thorium and actinium. The uranium and radium series can, in fact, be combined, for it seems to be proved that radium is a derivative of uranium. In the radium series the last known radioactive body is polonium, the production of which by radium is now a proven fact. It is likely that the actinium series is related to that of radium.

We have seen that helium gas is one of the products of radium disintegration. The helium atoms are detached from those of radium and its derivatives during the course of the transformation. It is supposed that after the departure of four atoms of helium, the radium atom yields one atom of polonium; the departure of a fifth helium atom determines the formation of an inactive body with an atomic weight believed to be equal to 206 (20 units below that of radium). According to Rutherford this final element is nothing more than lead, and this supposition is now being subjected to experimental

The relatively large amount of polonium prepared by Curie and Debierne has allowed an important study to be undertaken. This consists in counting a large number of alpha particles emitted by polonium and in collecting and measuring the corresponding volume of helium. Since each particle is a helium atom, the number of helium atoms is thus found which occupy a given volume and have a given weight. It can therefore allow us to deduce, in a general way, the number of molecules in a gram molecule. This number, known as Avogadro's constant, is of great importance. Experiments conducted on polonium have supplied a first value for this number, which is in good agreement with the values obtained by other methods. The enumeration of alpha particles is done by an electrometric method due to Rutherford; this method has been brought to perfection by means of a photographic recording apparatus.

Recent investigations have shown that potassium and rubidium emit a very feeble radiation, similar to the beta radiation of uranium and radium. We do not yet know whether we should regard these substances as true radioactive bodies, i.e. bodies in the process of transformation.

To conclude I should like to emphasize the nature of the new chemistry of radioactive bodies. Tons of material have to be treated in order to extract radium from the ore. The quantities of radium available in a laboratory are of the order of one milligram, or of a gram at the very most, this substance being worth 400,000 francs per gram. Very often material has been handled in which the presence of radium could not be detected by the balance, nor even by the spectroscope. And yet we have methods of measuring so perfect and so sensitive that we are able to know very exactly the small quantities of radium we are using. Radioactive analysis by electrometric methods allows us to calculate to within 1% a thousandth of a milligram of radium, and to detect the presence of 10^{-10} grams of radium diluted in a few grams of material. This method is the only one which could have led to the discovery of radium in view of the dilution of this substance in the ore. The sensitivity of the methods is still more striking in the case of radium emanation, which can be detected when the quantity present amounts, for example, to only 10^{-10} mm³. As the specific activity of a substance is, in the case of analogous radiations, approximately in inverse proportion to the average life, the result is that if the average life is very brief, the radioactive reaction can attain an unprecedented sensitivity. We are also accustomed to deal currently in the laboratory with substances the presence of which is only shown to us by their radioactive properties but which nevertheless we can determine, dissolve, reprecipitate from their solutions and deposit

Marie Curie

More

we'll call the chemistry of the imponderable.

*The lecture was held in the lecture hall at the Royal Academy of Sciences. The Royal Academy of Sciences were at the time located in the Westman Palace, a building behind Adolf Fredrik church in Stockholm, Sweden.

1. H. Becquerel, *Compt. Rend.*, (1896).
2. P. Curie and M. Curie, *Compt. Rend.*, (1898); (1899).
3. M. Curie, *Rev. Gen. Sci.*, (1899); *Rev. Sci.*, (1900).

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The Nobel Prize in Chemistry 1911

Marie Curie

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