



V. Actinium and its successive products

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In the silence of the authorities, I have myself suggested that Boltzmann's $e^{-2h\chi}$ law supplies the necessary guidance. The chance that any group or system of molecules shall be in a configuration in which the potential of their mutual forces, and of the external forces if any, is χ , is proportional to $e^{-2h\chi}$. Or, if you prefer so to state it, the time during which on the average of any very long time they will be in that configuration is proportional to $e^{-2h\chi}$. That gives the minimum χ for a statical system as a particular case. For if χ_0 be the potential in the configuration A_0 , and χ_1 in the configuration A_1 , and if $\chi_0 < \chi_1$, A_0 is more probable than A_1 in the ratio $e^{2h(\chi_1 - \chi_0)}$, that is in an infinite ratio in the statical system, for which h is infinite. The statical system must therefore be in minimum potential. Also if there be only external forces acting, Boltzmann's law gives $e^{-2h\chi}$ as the density at the point where the potential is χ , as in Maxwell's vertical column of air. I worked out the consequences of the application of the law to the general case in a former paper (Phil. Mag. for October 1901), and I think my conclusions were in the main right. If so, the law would be inconsistent with Lord Rayleigh's symmetry, and with its consequences. In fact it seems to me that Lord Rayleigh's symmetry and Boltzmann's law cannot both be true for one and the same system in the same state.

It may be said perhaps that Boltzmann's law holds only for external, and not for intermolecular forces. Some English writers, notably Dr. Watson, while not expressing their disagreement with the law as applied to intermolecular forces, prefer to let it alone. That I think arises from excess of caution, or perhaps because the law, if so applied, leads to results inconsistent with some favourite doctrines of the orthodox theory of gases. The proof of the law given by Boltzmann at p. 134 of his *Vorlesungen*, Part I., is formally applicable to intermolecular forces. Why may we not so apply it?

V. Actinium and its Successive Products.

By T. GODLEWSKI, Ph.D. (Cracow)*.

RUTHERFORD and Soddy, in their well-known investigations† on the activity of thorium, have shown that it is possible to separate from it a very active constituent

* Communicated by Prof. E. Rutherford, F.R.S. Presented before the Academy of Sciences in Cracow, April 3, 1905.

† Rutherford and Soddy, Phil. Mag. Sept. and Nov. 1902; Trans. Chem. Soc. lxxxi. pp. 321 & 807 (1902).

which they call ThX. The activity of this product decays with the time according to an exponential law; *i. e.*, the equation of monomolecular chemical reaction, falling to half value in about four days. At the same time, the thorium, which by the removal of thorium X had been deprived of about 75 per cent. of its activity, recovered its activity, the recovery curve being complementary to the curve of decay of ThX. The substance U_rX, discovered earlier by Crookes, acts in a manner analogous to ThX. The β activity of this substance decayed according to an exponential law with the time, falling to half value in twenty-two days.

Rutherford and Soddy have explained these phenomena on the supposition that the radioactive bodies are producing fresh radioactive matter at a constant rate, and that the activity of the matter so formed decreases according to an exponential law with the time. The discovery of these phenomena supplied the basis for the disintegration theory, which supposes that the atom of a radioactive body breaks up through a series of well-marked stages. The resulting products are quite distinct bodies, though they escape detection by chemical methods on account of the minute amount of the substance under investigation. Their existence is proved first of all by electrical measurements which allow us to make the quantitative investigations of the rate of change of these products.

On looking over the series of successive products arising from different radioactive bodies, striking similarity between the products of thorium and actinium is at once manifest. Thorium produces ThX, ThX the emanation, this gives rise to the active deposit which undergoes two further transformations, the first slow change being a rayless one, the other emitting all kinds of rays. Actinium in like manner produces an emanation which is transformed into an active deposit which undergoes two further changes, the first being a slow rayless change and the other a rapid change.

This analogy in the number and nature of the products pointed to the possibility* that there existed between the actinium and its emanation an intermediate product which bore the same relation to actinium that ThX bears to thorium. In a letter to 'Nature' (26 Jan. 1905) I gave the preliminary results of the investigation which proved the existence of this product.

Taking into consideration the similarity of actinium and thorium, I applied to the actinium the same method which

* See Rutherford, Bakerian Lecture: "The Succession of Changes in Radioactive Bodies," Phil. Trans. Royal Soc. ser. A, vol. cciv. pp. 190 & 204.

had been used by Rutherford and Soddy for the separation of ThX from thorium.

The experiments were made with the emanating substance of Giesel, which, according to numerous investigations*, has been found to contain the same radioactive constituent as the actinium of Debierne. The saturation current due to the α -ray activity of the products under investigation was measured with a quadrant electrometer of sensibility 120 divisions per volt; the needle was kept at the standard potential of 300 volts. The β activity was measured by means of a sensitive electroscope. Four different sets of experiments were made which gave very concordant results.

In each case 0.15 gr. of the emanating substance, of activity about 300 times that of uranium, was dissolved in 250 cm. of hydrochloric acid (about 8 per cent. concentration). The solution was evaporated on the water-bath to about 100 c.cm. When ammonia was added to the solution, a reddish-brown substance (probably hydroxide) was precipitated. The precipitate, collected on a filter-paper, was dried as quickly as possible, and then its activity was measured. The filtrate was then evaporated to dryness, and when ammonium salts were driven off by ignition a small amount of a brown-black residuum was left behind on the dish. On raising this to a red heat, the colour of the residue changed from black to white. This residue was intensely active compared with the weight. Immediately after the dish was cold the activity of the residue was measured and was found to decrease slowly with the time according to an exponential law. In the same period the actinium, which by precipitation had been rendered almost inactive, recovered its activity, the recovery curve being complementary to the curve of decay.

From analogy to ThX, which it so closely resembles in radioactive properties, the active substance separated from actinium will be termed Actinium X (AcX).

The results of one of the series of experiments are given in Table I. and graphically represented in fig. 1. In Table I. the first column gives the days measured from the time of separation; the second column gives the activity. The activity of AcX is expressed as a percentage of the initial activity, the maximum activity soon after separation being taken = 100. But for actinium (deprived of AcX) the final value is taken = 100.

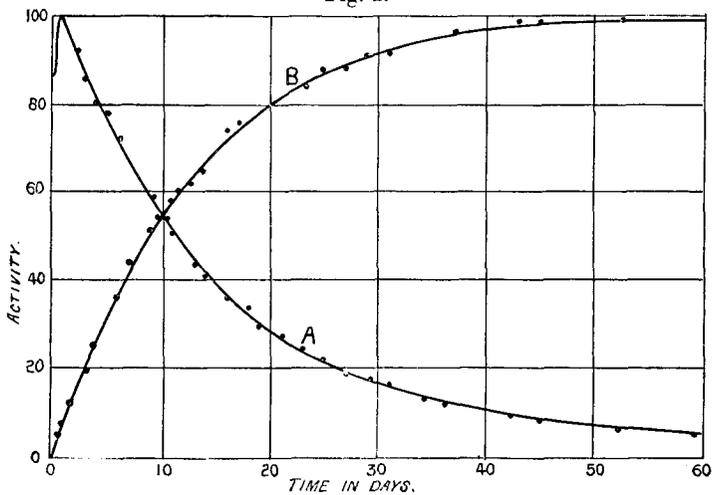
In fig. 1, curve A represents the activity of AcX as a function of the time, the activity of AcX being expressed in the same units as in Table I. In the recovery curve B the difference between the maximum and the first value is taken = 100.

* See f. i. Rutherford, Bakerian Lecture, *loc. cit.* p. 188.

TABLE I.

AcX.		Actinium.	
<i>t</i> in days.	Activity.	<i>t</i> in days.	Activity.
0.25	87.5	0.1	5.25
0.9	100	0.7	9.22
1.9	92.6	1.7	16.7
3.0	86.0	2.7	24.0
4.0	81.5	3.7	29.6
5.9	72.6	5.7	39.8
6.9	70.7	6.7	48.0
8.9	59.2	8.7	52.5
9.9	54.5	9.7	56.5
10.9	50.8	10.7	61.4
12.9	43.6	12.7	62.5
13.9	41.2	13.7	68.6
14.9	40.5	15.8	76.2
15.9	36.2	17.0	78.0
17.1	35.4	20.7	82.3
17.9	34.0	22.7	85.7
21.0	26.5	25.0	88.6
22.9	24.4	27.0	90.0
24.9	21.8	30.8	92.3
27.2	18.5	36.5	97.1
29.2	17.6	45.0	99.5
30.9	16.1	52.0	99.0
34.0	13.5	64.0	100
36.0	12.1	67.0	100
42.0	8.82		
45.0	7.77		
52.0	5.89		
59.0	4.63		
67.0	3.69		

Fig. 1.



The activity of AcX immediately after removal was weight for weight more than a hundred times as great as that of the original actinium. In the various experiments the value of the initial activity of AcX was proportional to the amount of actinium used, but was by no means proportional to the total weight of matter obtained from the filtrate. In some cases, for example, only a few milligrams of the substance were obtained, which exhibited as great activity as a few centigrams obtained in other cases. This shows clearly that in the case of actinium, the substance obtained from the filtrate, which we see and weigh, does not all consist of AcX. The substance contains some impurities; in the present case, probably some of the rare earths. The amount of actinium X actually present is so minute that it precludes the possibility of a direct chemical investigation of its properties.

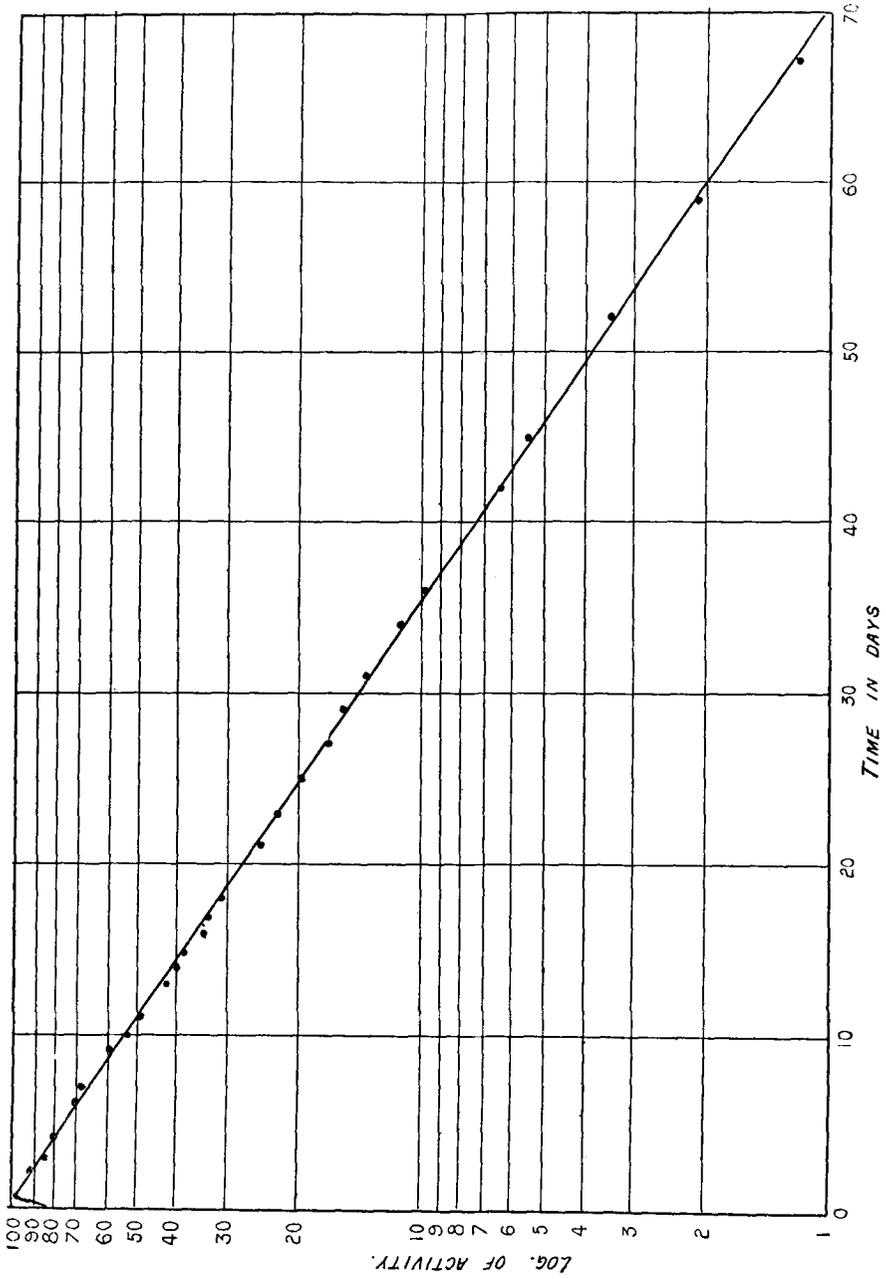
We see from Table I. that the activity of AcX increases in the first day after removal to about 15 per cent. of its original value, reaches a maximum, and then decays with the time according to an exponential law, falling to half value in 10.2 days. This exponential law of decay is clearly seen in fig. 2, where the ordinates represent the logarithms of the activity of the product AcX, and the abscissæ the time after separation. On subtracting from the quantities given in the Table I. the number 2.70, which represents the activity of the residue which did not decay with the time, the points fall accurately on a straight line, as in the figure. This non-decaying residue comes probably from the small amount of actinium, which is not precipitated, and is therefore present in the filtrate.

The activity of actinium, from which actinium X was removed, increased so that the recovery-curve was approximately complementary to the decay-curve of AcX. The small difference between the experimental and the theoretical curve, as expressed by the equation $I_t = I_0(1 - e^{-\lambda t})$, where λ has the same value as in the decay-curve, is probably due to a variation in the rate of escape of the excited activity, which is extremely volatile. In all cases the decay-curve agreed more closely with the theoretical equation than the recovery-curve.

The initial increase of activity of AcX immediately after removal (see fig. 1 A and fig. 2) is analogous to the similar increase of activity of ThX. The only difference is that the recovery-curve of actinium does not show the same initial decay as found in the case of thorium*. This fact is explained by the different properties of the excited activity

* See Rutherford, 'Radioactivity,' pp. 180 & 295.

Fig. 2.



of actinium and thorium. The active deposit of actinium is soluble in ammonia, and is volatile when heated*. The active deposit of thorium, on the other hand, is not soluble in ammonia and is not so readily volatilized. The initial increase of activity of AcX is thus explained in the following manner.

When actinium is precipitated with ammonia, the active deposit is left behind in the filtrate together with AcX. In the moment, however, when we heat, the volatile active deposit is driven off also. But as soon as AcX is separated, it at once produces the emanation which gives rise to the active deposit. The activity of the latter, at first, more than compensates for the decay of activity of AcX, which has a comparatively slow change, and in consequence the activity of AcX first of all increases.

On the other hand, the actinium, when treated with ammonia, was deprived not only of AcX but also of most of the active deposit. Any of the latter, if still remaining in the precipitate, would be driven off during the process of drying. In consequence, when we start the measurements of the activity of the precipitate itself, no excited activity is present. The activity at once commences to increase, since a fresh amount of AcX is produced which in turn gives rise to excited activity. In consequence we do not observe the initial decay in the recovery-curve of actinium as in the corresponding curve for thorium.

Disregarding these small peculiarities, the behaviour of the product AcX, and of actinium deprived of AcX, is, as we have seen, completely analogous to that of ThX and of thorium deprived of ThX. There is, however, the following distinct difference. After removal of ThX, thorium always has a certain amount of residual activity about 25 per cent. of the maximum value. A similar effect is observed in the case of radium, where the "de-emanated" radium has always a non-separable activity of about the same (25 per cent.) value. In the case of actinium, immediately after removal of AcX the actinium is almost inactive, its activity being only 5 per cent. of its maximum value. I tried experiments to see whether this activity could not be removed by means of successive precipitations with ammonia, but although eight precipitations were made in the course of seven hours the residual activity always remained. Nevertheless, the smallness of the initial amount of activity pointed to the probability that in reality

* The more complete account of the physical and chemical properties of the active deposit of actinium will be published later, as the experiments are not yet completed.

the actinium itself is not active, and that the residual activity observed is due to a small quantity of AcX, which is left behind. The interval between the last precipitation and the first measurement was always one hour or more, but this alone would not account for the observed current. It seems very probable that at the moment of the removal of AcX, *if the separation were complete*, actinium would be entirely devoid of activity. From the point of view of the theory of radioactive changes, this shows that the change of actinium into AcX is not accompanied by either α , β , or γ rays or, in other words, is a "rayless" change.

By means of an electroscope, it was found that actinium X gave out all three kinds of rays α , β , and γ . Now the products of excited activity are very quickly formed owing to the very rapid change of the emanation. The activities of these products are consequently measured together with AcX. It was separately proved that the active deposit gave out β rays*. Taking into consideration the analogy with thorium and even with radium, we should expect that the measured β activity of AcX arises not from AcX itself, but from the excited activity resulting from it. There is, however, strong evidence that in the case of actinium the β and probably the γ rays are emitted also by AcX itself. For instance, the curves of decay of activity of AcX measured by α and β rays are throughout identical, even from their beginning; and further, the activity of AcX measured by β rays, five minutes after strong heating, when all the volatile excited activity should be driven off, exhibits a very great initial value, which could not be the case if the rays were emitted only by excited activity.

It is thus most probable that AcX itself gives rise to all three kinds of rays.

Source of the Actinium Emanation.

In the case of thorium the product ThX was intermediate between thorium and its emanation. In order to see whether AcX occupies the same position in actinium, I measured the rate of change with time both of the emanating power of AcX and of actinium from which AcX was removed. The measurements of activity of the emanation were made in a cylindrical brass testing-vessel †, in the interior of which three insulated

* It was found that the excited activity of actinium, measured by β rays, after a long exposure decayed according to an exponential law with the time, falling to half value in 36 minutes. The complete account of these investigations will be given in another place.

† See Rutherford, 'Radioactivity,' p. 199, and fig. 37.

electrodes were placed; during the measurements one of the electrodes was connected with an electrometer and the other two were earthed.

Both actinium X and actinium deprived of AcX were placed in solutions of ammonium chloride in two bubbling flasks, and these could be successively connected with the testing cylinder in which the amount of emanation was measured. For the purpose of comparison with a substance of a standard emanating power, the following arrangement was used.

The current of air passed through the bubbling bottle and carried with it the emanation of the product to be investigated. It thus passed through the testing cylinder, in which the activity of the emanation was measured. On leaving the cylinder, the current of air entered a glass tube 2 cms. in diameter and about 3 metres long. At the end of this glass tube some fresh solid actinium was placed, and the emanation from this was carried into the second testing cylinder where its activity was measured.

In this manner the same current which carried the emanation from the product under investigation also carried the emanation from the standard actinium. In passing through the long and wide glass tube the emanation which left the first cylinder decayed completely before reaching the second cylinder. By this method of measurement, the emanating power of AcX was directly compared with the standard emanating power of solid actinium.

The experiments made in this manner showed:

- (1) That the actinium immediately after removal of AcX gives practically no emanation.
- (2) That the rate of increase of the emission of emanation of actinium after removal of AcX is the same as the rate of increase of its activity.
- (3) That the emanating power of AcX decreases at the same rate as the activity of AcX.

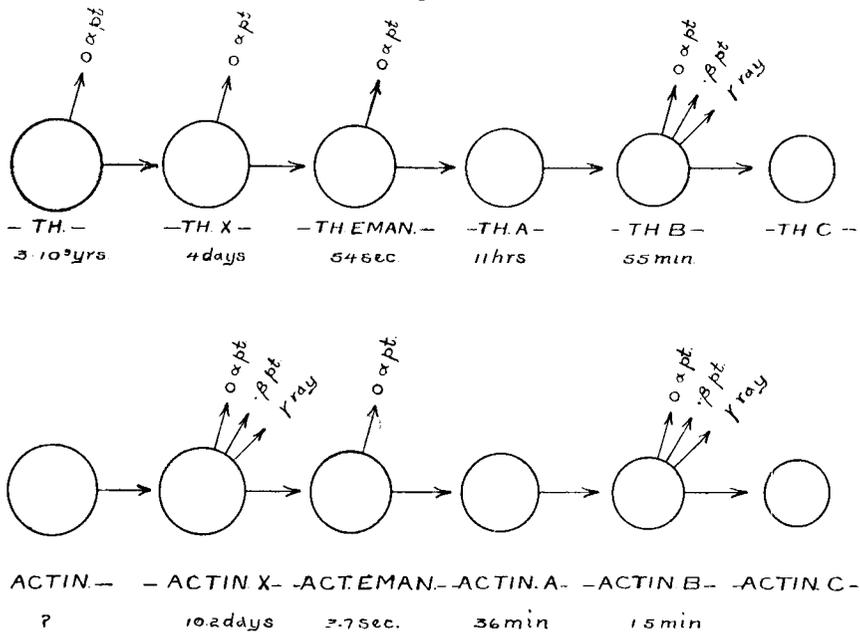
Since the emanation is only observed when actinium X is present, and is always proportional to the amount of actinium X, it must be a product of actinium X.

The changes occurring in actinium are shown in the following graphical representation*, together with the period required for transformation to half value. For comparison the changes taking place in thorium are also given.

* See Rutherford, Bakerian Lecture, *loc. cit.* pp. 180-190 & 204; also Miss Brookes, *Phil. Mag.* Sept. 1904, pp. 382-384; also Brouson, *Amer. Journ. of Science*, vol. xix. Feb. 1905, p. 187.

It is seen that there is a very striking similarity between the number and nature of the changes for actinium and thorium. But the periods of decay, the radioactive, chemical

Fig. 3.



and physical properties of the products of actinium all point conclusively to the fact that we have in actinium a distinct chemical element.

Whilst writing this paper, the December number of the *Jahrbuch für Radioaktivität und Elektronik* was received containing a paper by Giesel on Emanium*. In this paper Dr. Giesel gives an account of his investigation, in which he finds that it is possible to separate from emanium by precipitation with ammonia a small amount of very strongly active substance. The method of separation employed by him was then identical with the method I have used. I cannot, however, compare quantitatively my results with his, inasmuch as Dr. Giesel does not publish any measurements.

A short account will now be given of some experiments which are still in progress on the nature of the β and γ rays of actinium.

* Giesel, "Untersuchungen über das Emanium (Actinium)," *Jahrbuch f. Radioakt.* vol. i. pp. 375-358.

The β rays of actinium are completely distinct in their character from the β rays emitted by the other radio-elements, inasmuch as they are completely homogeneous. This fact was established by the measurements of absorption of β rays in passing through solid bodies. The activity measured by β rays decreased strictly according to an exponential law with the thickness of matter traversed. The equation $I = I_0 e^{-\lambda d}$, where d is the thickness, was applicable even in the case when I was less than 1 per cent. of its original value.

The β rays from actinium differ also from the β rays of other radioactive elements in the absolute value of the absorption constant λ , which is about 2.5 times as great with actinium as with uranium. Thus the β rays of actinium have less than half the penetrating power of those emitted by any other radio-element.

The existence of the γ rays from actinium was also distinctly proved. The absorption measurements showed that the γ rays of actinium are fairly homogeneous, and their penetrating power was only about one quarter of that observed for the rays from radium. A more complete account of these investigations will be published later on.

In conclusion it is my most pleasant duty to express my deepest gratitude to Prof. Rutherford for suggesting these investigations, for his kindness in the advice he has so freely given to me, and for placing at my disposal all the plentiful resources of his laboratory at McGill University.

McGill University, Physics Building,
February 24, 1905.

VI. *Some Radioactive Properties of Uranium.*

By T. GODLEWSKI, *Ph.D.* (Cracow)*.

1. *The Discovery of UrX.*

IN 1900 Sir William Crookes † showed that it is possible to separate from uranium by a single chemical operation a small amount of radioactive substance to which he gave the name UrX. This substance was, weight for weight, many hundred times more active photographically than the uranium from which it had been separated. The uranium deprived of this substance was almost inactive.

Similar results were afterwards observed by Becquerel ‡,

* Communicated by Prof. E. Rutherford, F.R.S. Presented before the Academy of Sciences in Cracow at the sitting of 9th of May, 1905.

† Crookes, Proc. Roy. Soc. lvi. p. 409 (1900).

‡ Becquerel, C. R. cxxxi. p. 137 (1900); cxxxiii. p. 977 (1901).