LXXXV.—*The Radioactivity of Uranium.*

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DURING the course of the investigation contained in the preceding paper on the radioactivity of thorium, it became advisable to investigate in parallel a radioactive element that does not give out an emanation or excite radioactivity on surrounding bodies. It has been shown that these secondary actions always form a possible factor in the results obtained with thorium. For this purpose, the radioactivity of uranium was chosen, but at the outset great differences arose between the results obtained and those published by prior investigators. These differences led to a comparative study being made in certain cases of the photographic and electrometer methods of measuring uranium radioactivity, which result in an explanation of the anomalous results that had been obtained.

Sir William Crookes (*Proc. Roy. Soc.*, 1900, 66, 409) made the

remarkable discovery that in one chemical operation uranium could be obtained inactive to the photographic plate, whilst the whole of the activity was concentrated in a small non-uranium residue. This residue, to which he gave the name UrX , was at least a hundred times more active than the uranium from which it had been separated.

On repeating this separation, which consists in precipitating solutions of uranium with ammonium carbonate and redissolving the precipitate in excess of the reagent, it was found that the small residue of UrX was almost inactive, whilst the uranium contained in the ammonium carbonate filtrate, possessed about the normal amount of activity. The electrical methods of Professor Rutherford were employed for measuring radioactivity. The same negative result was obtained by the other method of separation employed by Crookes. If crystallised uranium nitrate is dissolved in ether, the uranium divides itself in two unequal fractions between the ether and water present. That dissolved in the ether layer, which is the major fraction, was found by Crookes to be inactive to the photographic plate, whilst the small part dissolved in the aqueous layer possessed all the activity contained in the original uranium nitrate. As in the former case, however, on repeating the operations, both fractions appeared almost equally active to the electrometer, and no separation had apparently been effected.

The same preparations were then examined for their action on the photographic film, and gave results exactly the converse of those observed with the electrometer and in accord with the published results of Sir William Crookes.

Rutherford (*Phil. Mag.*, 1899, [v], 47, 109) showed that uranium radiation consists of two different types which he named α and β . The α -radiation is absorbed very readily even by gases, being reduced to half its value by passage through 4.3 mm. of air. The β -radiation, on the other hand, is very penetrating in character, and is but little absorbed by gases. It is able to pass through 0.5 mm. of aluminium before it is reduced to half its value. The latter part constitutes only a few per cent. of the total radiation of the uranium. In addition, by the electrometer method of measurement, under ordinary circumstances, a fraction only of the intensity of β -radiation is recorded, for this method depends on the ionisation of the air, and this is proportional to the absorption of the rays by the air. It follows therefore that under ordinary circumstances, in which the rays traverse a few cm. only of air, the electrometer method measures practically the α -radiation of uranium alone. The photographic method employed by Crookes, in which the rays are made to pass through glass or card before reaching the sensitive film, will, on the other hand, measure only the β -radiation. Hence the results obtained would be explained if

the methods employed separated the β -radiation only, and left the α -radiation intact.

Becquerel has shown (*Compt. rend.*, 1900, 130, 1584) that the rays from uranium are deflected in the magnetic field, and therefore consist of cathode rays. Rutherford and Grier, *Phys. Zeit.*, 1902, 3, 385), in a general investigation of the rays from various radioactive substances to determine what proportion in each case was deviable in the magnetic field, and therefore cathodic in nature, had already shown that the β -radiation of uranium is entirely cathodic, and the α -radiation not at all. They undertook the examination of the specimens that had been prepared in the course of the present work, the details of which appear in their paper, with the special apparatus employed. It suffices to state here that in the arrangement they adopted the rays are made to pass through a sufficient thickness of air to absorb a considerable part, and the sensitive Dolezalek electrometer was employed. Under these circumstances, UrX was found to be giving out ionising rays abundantly. This radiation was shown to be the β -radiation of uranium only, practically free from α , for it passed through aluminium foil without great loss, and is deviable to the same extent as the β by the magnetic field. The rays from the photographically inactive uranium were found, on the other hand, to consist entirely of α -radiation and gave no rays deviable in the magnetic field or capable of passing through thin aluminium foil. I must here express my great obligation to Professor Rutherford and Mr. Grier for permission to use these results.

It thus appears that the methods of chemical separation employed by Crookes effect a separation only of the constituent responsible for the β -radiation. The uranium after the process still retains the whole of the non-deviable radiation, which in the electrometer method contributes the greater part of the effect. In this respect, uranium is analogous to thorium, as shown in the preceding paper (p. 854). The excited radioactivity produced by ThX as a secondary effect, which itself comprises both deviable and non-deviable radiation, makes it impossible at present, however, to say whether the primary radiation of ThX is, like that of UrX, wholly cathodic or not. The non-separable parts in both cases consist entirely of rays non-deviable in the magnetic field.

The next point to be decided was whether the α -radiation of uranium could affect the photographic plate. A sample of the preparation, free from β -radiation, was exposed about 5 mm. away from a sensitive film for 72 hours without an intervening screen. Only a very slight darkening occurred, so that the action, if any, of the α -radiation may be considered negligible in comparison to the effect of the β -radiation. It is not possible to try longer exposures than 3 days by this method,

owing to the regeneration of the β -radiation, which will be referred to later. However, Becquerel (*Compt. rend.*, 1902, 134, 208) exposed plates for 20 and 49 days near uncovered uranium salts in a magnetic field where the β -radiation would be deflected and eliminated, and failed even after these long periods to obtain an impression on the sensitive film. He concluded that the whole of the uranium radiation is cathodic, and can be deflected by the magnetic field. The foregoing considerations make it clear that the α - or non-deviable radiation of uranium, which contributes the major part of the ionisation effect, is without appreciable action on the photographic plate.

In the light of these results, and those obtained for thorium in the preceding paper, the method employed by Becquerel (*Compt. rend.*, 1900, 131, 137) to separate the active constituent of uranium is of interest. Becquerel precipitated barium as sulphate in solutions of uranium, and found that after successive precipitations the activity of the latter was much enfeebled, both towards the electrometer and the sensitive film. As, however, he enclosed the salt in paper for the electrometer measurements, and this absorbs the α -radiation almost, or quite, completely, the result must not be taken to mean that both the α - and the β -radioactive materials were separated by the process. On repeating the separation, it was found that, as in the case of the methods of Crookes, the β -radioactive constituent only was removed. After four successive precipitations in one day, the β -radiation was found to be reduced to 8 per cent. of its original value, and eight more precipitations on the day following completely removed it. But the activity of the resulting uranium preparation to the electrometer under ordinary circumstances was hardly appreciably decreased. The α -radiation had therefore not been affected.

Becquerel has already shown (*Compt. rend.*, 1901, 133, 977) that the uranium after this treatment recovered its normal activity on standing. With the sensitive electrometer, the presence of cathode-rays in a product originally quite freed from them can be detected after three days. It thus appears probable that what has been shown to hold true for ThX applies equally to UrX, and the subject is under investigation conjointly with Professor Rutherford. The question already discussed for thorium therefore arises in connection with the non-separable activity of uranium:

1. Is this residual activity to be regarded as a secondary radiation produced by the presence of UrX? Or,
2. Is it caused by a distinct material substance capable of chemical separation?

On the first view, if UrX—the cause of the phenomenon—is removed, the residual activity will decay with time. The following experiment was therefore performed. The solution of uranium nitrate, which had

been successively precipitated with barium sulphate twelve times, and had been shown to be free from β -radiation, was allowed to stand. Every three or four days it was once more precipitated in the same manner, and the UrX produced during that period removed from the uranium. After twenty-one days, the uranium in the solution was precipitated with ammonia and converted into the green oxide. For comparison, a sample of the same uranium nitrate, as obtained from the maker, was subjected to two precipitations with barium sulphate to remove some of the β -radioactive constituent, the uranium then being precipitated and converted into oxide as in the former case. The radiations from the two samples were then directly compared. In the one, the α -radiation had been given three weeks to decay, and by comparison with the other a direct measure of the diminution suffered during this period could be obtained. Not the least difference, however, could be detected in the intensities of the radiations in the two cases. The actual values obtained were within 1 per cent. of each other. If it is assumed that in this experiment a difference of 5 per cent. could be with certainty detected, the conclusion is reached that if the α -radiation of uranium is a secondary phenomenon produced by the β , it takes at least a year to decay to half value. As this is not in accordance with what is known of the nature of excited radioactivity, the view cannot be regarded as probable.

On the second view, the α -radiation is produced by a second distinct type of matter. As in the case of thorium, however, all attempts made to separate such a constituent of uranium by chemical methods have so far failed, although the methods have not yet been exhausted. It is however, interesting to note that polonium, discovered by Curie, fulfils in almost all respects the functions of this hypothetical constituent. It gives only non-deviable radiation, and Professor Rutherford, to whom the suggestion is due, found that the penetrative power of the rays from polonium is very similar to that of the uranium radiation. Moreover, M. and Mme. Curie (*Compt. rend.*, 1902, 134, 85) have stated that its activity slowly decays with time, which is to be expected *after*, but not before, it has been separated from the uranium producing it. However, experimental work has not yet justified this suggestion.

The actinium of Debierne, according to a recent paper by Crookes (*Chem. News*, 1902, 85, 109), has been found to be identical with UrX although no reference is given.

In the paper just quoted, Crookes brings forward some results obtained by the photographic method which led him to the conclusion that UrX gives a radioactive emanation similar to that given by thorium and radium. No evidence of such a radioactive emanation has been obtained in the course of the present work, even with th

most sensitive electrometer, and it appears probable that the cause of the darkening of the photographic plate by this substance in the cases where the film is shielded from direct radiation is not a radioactive substance in the accepted sense of the word, but an agent similar to hydrogen peroxide in its photographic actions.

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