Products of the Uranium Fission. Radioactive Isotopes of Iodine and Xenon*

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Radioactive iodines resulting from the uranium fission have been further studied, with results which in addition to confirming those of previous investigators, establish the following active substances as products of the uranium fission.

$$I^{>131} \xrightarrow{6.6\text{ hr.}} Xe^{>131} \xrightarrow{9.5\text{ hr.}} Cs\ (?)$$
$$Xe^{>131} \xrightarrow{4.3\text{ day}} Cs\ (?)$$

The 4.3-day xenon is produced by the decay of the 22-hour iodine. The 6.6-hour iodine is either a direct fission product, or the daughter of an active tellurium of half-life less than 1 minute.

I. INTRODUCTION

SINCE the discovery by Hahn and Strassmann[1] that radioactive barium isotopes result from the neutron irradiation of uranium, many other active substances have been found to be produced in the same way, and have been studied by numerous investigators.[2] Radioactive isotopes of most of the elements from selenium to lanthanum have been reported. The chemical identities of the active substances have been established principally by characteristic chemical reactions, and in a few cases by critical absorption measurements on characteristic x-radiations emitted by the active bodies.

Shortly after the discovery that the 2.5-hour "transuranium" is actually an iodine isotope[3,4] we looked for other radioactive halogens in neutron-irradiated uranium and found active iodines to which we ascribed half-lives of about 45 minutes and about 12 hours.[5] A detailed study by Abelson[6] showed that radioactive iodines of half-lives 54 minutes, 2.4 hours, 22 hours, and 8 days are formed, all descended from active telluriums. These substances were also studied by Hahn and Strassmann[7] with essentially the same results except that a half-life of 18.5 hours instead of 22 hours was assigned to one of the iodines.

We have continued our investigation of these substances with results which now stand in agreement with those of Abelson, and which also show the formation of an iodine of 6.6-hour half-life, for which no tellurium parent could be found, the growth of an active xenon (half-life 9.5 hours) from this iodine, and the formation of a 4.3-day active xenon, probably from the 22-hour iodine. In the course of this work, we have learned of unpublished investigations by Segrè, Langsdorf, Jr. and Wu,[8] apparently carried out along similar lines, and with results in substantial agreement with ours, which we report in the following sections.

II. THE ACTIVITY SEPARABLE WITH IODINE

Iodine separated from neutron-irradiated uranium had an activity whose decay curve clearly shows the presence of at least four active substances. A typical decay curve for such a separation is given in Fig. 1. The uranium was irradiated for 30 minutes; the separation was completed 15 minutes after the end of irradiation, which was taken as the zero of time. The measurements were discontinued before the 8-day component was resolved, and the curve drawn for the 8-day component was calculated from the slope near the end of the experimental curve, under the assumption that only the 22-hour and 8-day

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[1] O. Hahn and F. Strassmann, Naturwiss. **27**, 11 (1939).

[2] These are cited in detail by L. A. Turner, Rev. Mod. Phys. **12**, 1 (1940).

[3] P. Abelson, Phys. Rev. **55**, 418 (1939).

[4] N. Feather and E. Bretscher, Nature **143**, 516 (1939).

[5] R. W. Dodson and R. D. Fowler, Phys. Rev. **55**, 880 (1939).

[6] P. Abelson, Phys. Rev. **56**, 1 (1939).

[7] O. Hahn and F. Strassmann, Naturwiss. **27**, 529 (1939).

[8] Cited by J. J. Livingood and G.T. Seaborg, Rev. Mod. Phys. **12**, 30 (1940).

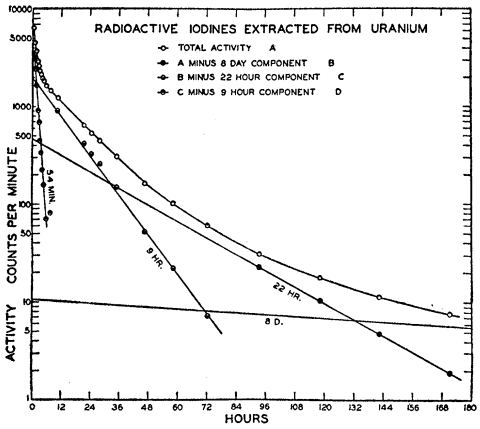

FIG. 1. Decay curve of the activity extracted with iodine from neutron irradiated uranium, and its analysis into various components.

components were present. Subtraction of this curve from the experimental points gave points (*B*), which lie on a curve terminating in a 22-hour slope, and successive subtractions gave curves (*C*) and (*D*), of slopes corresponding to 9-hour and 54-minute half-lives, respectively. No 2.4-hour contribution appears in this analysis, presumably because the yield of its long lived (77-hour) parent tellurium was so small and the complexity of the curve is so great that it could not be resolved. Three of the half-lives agree with those reported by Abelson;[6] in addition, it is seen that at least one additional active substance of half-life about 9 hours is present. The points (*C*) lie above the middle portion of the 9-hour line, and the best fit would be obtained with a curve concave downwards. Although this effect is small, it was checked in duplicate runs, and hence we suspected the presence of a radioactive daughter, xenon, growing from an active iodine. Subsequent experiments showed that two active xenons are formed. Hence the analysis of the total decay curve in Fig. 1, which ignores growth of active daughter substances, is not correct. However, because an unknown fraction of the active xenons escapes from the sample during measurement, and because of the probable difference in the efficiency of counting the beta-particles from the parent iodine and the daughter xenon, we were unable to draw a theoretical curve for the activity due to the parent-daughter combination, and resorted to other means of determining the half-lives of the active substances in question.

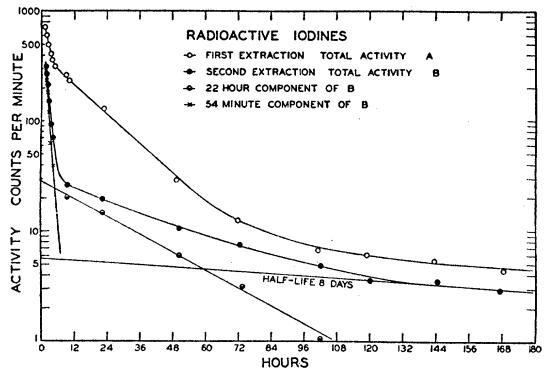

FIG. 2. Decay curves of two successive iodine extracts from the same sample of neutron irradiated uranium, and analysis of the decay curve of the second extract.

These effects make the final difference curve (for the 54-minute iodine) of only semi-quantitative significance, showing that an activity of about one-hour half-life is present.

Since Abelson's studies of the iodines resulting from the decay of the active telluriums from the uranium fission showed no evidence of any 9-hour substance, it is reasonable to conclude that any tellurium parent of this substance must have a very short half-life. This was confirmed by successive iodine extractions in the following experiment.

The uranium solution was irradiated for ten minutes, such a short irradiation time being employed in order to enrich the yield of any direct fission product, or one having very short-lived progenitors, over that of products descended from relatively long-lived progenitors. Extraction of iodine from the solution was completed 5 minutes after the end of the irradiation, special care being taken to insure a quantitatively complete separation. The uranium solution then stood for 15 minutes, and a second iodine extraction was made. The two iodine extracts were examined for activity as usual. The results are shown in Fig. 2. The first extract shows a decay curve similar to that in Fig. 1; the presence of the "9-hour" component in considerable amount (about 400 counts per minute at time zero) was confirmed by analysis of the curve (not shown in the figure). The analysis of the decay curve of the second extract (shown in the figure) shows only the 54-minute, 22-hour, and 8-day iodines. Thus, any active parent of the 9-hour

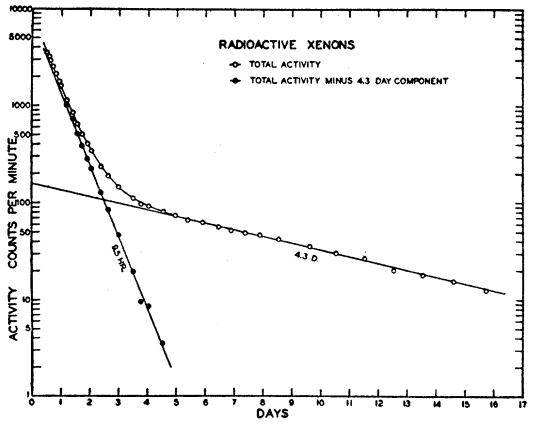

FIG. 3. Decay curves of the radioactive xenons separated from the active iodines.

substance must have disappeared (within experimental error) during the 5 minutes required for the first extraction. A 9-hour activity of 10 counts a minute could certainly have been observed; therefore at least 98 percent of the hypothetical parent must have decayed in the five minutes required for the first extraction, and consequently its half-life must be less than one minute.[8a]

III. Active Xenons Separable from the Iodine Extracts

Active xenons with half-lives of the order of a minute or less, and about 15 minutes have been found in neutron-irradiated uranium by Hahn and Strassmann,[1, 7, 9] and others, by a gas-stream technique in which the noble gas was swept out of solution by a gas stream and trapped in suitable adsorbents, which were examined for the presence of active descendents (cesium, barium, lanthanum) of the xenon. Because of the downward concavity of the "9-hour" component of the iodine separation we inferred that active xenons might be growing in the silver iodide precipitate. An attempt was made to expel the xenon from a solution of active iodide into thin-walled glass bubbles. A feeble activity, too weak for accurate measurement was found. Recourse was then had to the ingenious method described by Langsdorf, Jr., and Segrè[10] for the preparation of emanating samples of bromine and iodine. The iodine was absorbed on silver-nitrate-impregnated silica gel from carbon tetrachloride solution. The silica gel was dried, and placed in a small glass bulb which was attached through a stopcock to a counter. The arrangement was such that no beta-particles from the active iodine could reach the counter, and emanated xenon could diffuse into the counter if the stopcock was open. To the bulb was also attached a second stopcock through which the system could be evacuated and refilled with argon. After installation of the emanating sample, the background of the counter was measured with the first stopcock closed. The cock was then opened, and there was a rapid growth of activity, attributable to diffusion of active xenon into the counter. Fig. 3 shows the decay of the active xenon collected over a growth period of 17 hours. Two active substances are present, of half-lives 9.5 ± 0.4 hours and 4.3 ± 0.4 days. Subsequent experiments proved that the entire activity could be removed by evacuating the counter, that the 9.5-hour substance is the daughter of a 6.6-hour iodine, and indicated that the 4.3-day activity is the daughter of the 22-hour iodine. We therefore attribute both activities to isotopes of xenon.

Because of the weak intensity of the longer lived xenon, we followed its activity for only about two half-lives after the 9.5-hour activity had become negligible. In the last measurement the activity was 12.5 counts per minute, or about half the background count as determined at the beginning of the run. When the background cannot be checked for small drifts in the course of a run (as in the present case), the measurement of such small activities is uncertain, and it was not considered profitable to carry the measurements further. The assignment of a 4.3-day half-life assumes, of course, that there were no appreciable drifts in the background, and that no other active substances were present.

[8a] *Note added in proof.*—Since the above was written, a letter has appeared by E. Segrè and C. S. Wu [Phys. Rev. **57**, 552 (1940)] on the subject of the active xenons produced in the uranium and thorium fissions. The results agree well with those herein reported, except that the parent of the 6.6-hour iodine is stated to be a tellurium of about 15 minutes half-life. Also, they report that one or more short-lived xenons grow from iodine.

[9] O. Hahn and F. Strassmann, Naturwiss. **27**, 163 (1939).

[10] A. Langsdorf, Jr. and E. Segrè, Phys. Rev. **57**, 105 (1940).

IV. The Iodine Parents of the Active Xenons

The genetic relationships between the active xenons and iodines were investigated by the technique of periodic separation of the daughter substances. The emanating-sample counter set-up, as described above, was used. The counter assembly was evacuated, then filled with argon to 5 cm pressure. Both stopcocks were immediately closed, and the background count was measured. The communicating stopcock between the emanating sample and the counter was then opened, and the growth of activity followed for about 7 hours. The counter assembly (including the bulb containing the emanating sample) was then evacuated, and the process repeated. The background count after each evacuation and refilling was constant during the course of the run, showing that the xenons left no active deposit, and thus that the 4.3-day substance is an isotope of xenon, rather than a radioactive cesium produced by the decay of the 9.5-hour xenon.

The activity of a daughter substance after growth for any fixed interval is proportional to the activity of the parent at any fixed time in that interval. In this case the xenon activity after growth for six hours was determined from the growth curves, and plotted as ordinate against the time (end of growth interval) as abscissa. The resulting curve, given in Fig. 4, is a decay curve for the iodines which decay to active xenons. It is seen that there is a major component of about 7-hour half-life, also that a longer lived component is present. Because of the very weak activity of the long-lived component, we were unable to follow its decay. However, we infer it to be the 22-hour iodine for the following reasons. The half-life is greater than 7 hours, and since the only such iodine activities occurring in appreciable yield are the 22-hour and 8-day iodines, we may conclude that the long-lived component is one of these. By comparing the data plotted in Figs. 3 and 4 the expected activity after the last 6-hour growth interval in Fig. 4 was calculated for each of the two cases. On the first assumption the expected activity was 2.5 counts per minute, on the second, 30 counts per minute. The measured activity was 2.0 counts per minute. We may therefore conclude that the 22-hour iodine is the parent of the 4.3-day xenon. Subtraction of a 22-hour curve drawn through the last point in Fig. 4 gave the difference curve in the figure, showing a pure exponential decay over a range of 1500-fold in the activity, with a half-life of 6.6 ± 0.3 hours. When a second iodine extraction was made from the uranium, 2.5 hours after the first extraction, the emanating sample prepared from the second extract gave only a weak activity, due to the growth of the 4.3-day xenon from the 22-hour iodine. This agrees with the observation that the 6.6-hour iodine does not continue to grow in the uranium for any appreciable time after irradiation. We therefore conclude that the 9.5-hour xenon is produced by the decay of an iodine of 6.6-hour half-life.

V. Discussion

In our previous experiments[5] on the active iodines from the uranium fission we were unable to follow the activity for more than about 50 hours because the counters used had relatively high backgrounds. Examination of the total decay curve in Fig. 1 shows that the complex of the longer lived components of the total activity closely simulates a 12-hour slope over this range of time. This effect explains the inconsistency of our preliminary report. The report of Hahn and Strassmann[7] of 18.5 hours instead of 22 hours for the half-life of one of the iodines is also significantly low. Since their measurements were on iodine separated directly from the uranium, which therefore contained the 6.6-hour iodine, it seems clear that their result has the same interpretation.

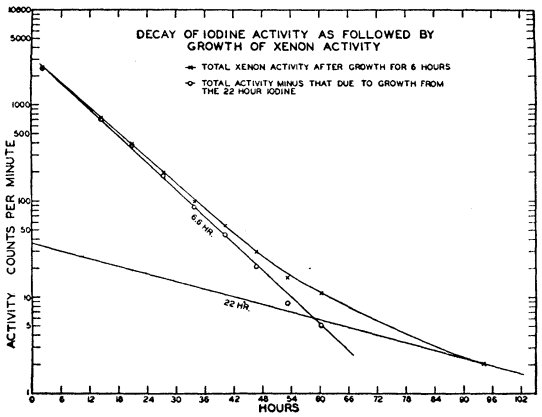

FIG. 4. Decay curve of the iodines which decay to active xenons, as followed by the growth of xenon activity.

In view of the great complexity of the total iodine decay curve, the values of the half-lives as determined by its analysis in Figs. 1 and 2 are not advanced as precise determinations of these quantities. The principal significance of the curve analyses in these figures is first, the agreement with the half-lives obtained by Abelson in his thorough study of the active iodines resulting from the decay of tellurium isotopes, and second, their demonstration of the presence of the 6.6-hour iodine formed either as a direct fission product, or as the daughter of a very short-lived tellurium.

The half-lives of the active xenons and their iodine parents herein reported do not correspond to those of any iodine or xenon isotopes of known mass (e.g. as determined by cross bombardments) so that we are unable at present to assign mass numbers to these substances. Since no cesium activity was found to grow from either of the xenons, the mass number of one of them may be that of stable cesium, 133; however, a very long-lived cesium would have gone undetected, and one of extremely short half-life would probably also have been unobserved. The known isotopes of iodine and xenon[8] from mass number 126 to 131 are stable or have half-lives different from those in question, so that we tentatively assign a mass number >131 to these active substances.

VI. Experimental Details

Solutions of uranyl chloride, of approximately 1 liter volume, and containing 1–2 moles of uranium, were bombarded with neutrons from the *D–D* apparatus in this laboratory. Total beam currents were 0.5–1.0 ma, at 250 kv; a heavy ice target was used. The brass can containing the ice target was covered with a thin film of paraffin and immersed in the uranium solution during bombardment. The solution was surrounded externally with several inches of paraffin.

For the separation of active iodines, about 15 mg of iodine in aqueous solution were added to the uranium and extracted by shaking with carbon tetrachloride, or carbon disulfide. The extracts were washed with three successive portions of water, in order to remove water-soluble contamination, and the iodine was reduced by shaking with water containing sodium bisulfite. To this aqueous solution was added sodium bromide, and sulfuric acid; the iodine was then quantitatively separated from bromine by oxidation with sodium nitrite, followed by extraction in carbon tetrachloride. The extracts were washed with water, the bisulfite reduction repeated, and the iodide precipitated with silver nitrate in the presence of nitric acid. The precipitate was collected on a 1-inch filter paper in a Hirsch funnel, washed with water, acetone, sucked dry, and examined for activity. Blank runs made in the same way on unirradiated uranium gave zero activity in the iodide precipitates.

For separation of active xenons, the above procedure, up to the second bisulfite reduction, was performed, and an emanating sample of the active iodine was prepared as previously described.

The iodide activities were measured with a ball-point counter, having a flat mica window, $1\frac{1}{8}$ inches in diameter, thickness, 2.9 mg/cm^2. The filter papers were mounted flat on glass plates, which could be placed about $\frac{1}{8}$ inch below the mica window in an exactly reproducible position. The counter was surrounded with about 2 inches of lead on all sides, and gave a background count of about 10 per minute. A high speed scale-of-8 circuit of the type described by Lifschutz[11] was used with the counter. During long runs the background count was repeatedly determined, and the counter sensitivity was checked for constancy with a standard uranium source of beta-particles. The sensitivity remained constant, and the background count did not vary by more than about 1 count per minute. The activities were measured by noting the time interval for 2400–4800 counts. As a result of these precautions, we believe the activity measurements to be reliable down to very low counting rates. Inspection of the decay curves shows that no large erratic fluctuations occur down to activities of a few counts per minute.

In measuring the xenon activities, it was not possible to observe all the above precautions, since the active material was inside the counters, and since the emanating sample assembly made lead shielding inconvenient. However, no signifi-

[11] H. Lifschutz, Rev. Sci. Inst. **10**, 21 (1939).

cant erratic fluctuations seem to be present in the measurements.

ACKNOWLEDGMENT

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Anomalous Scattering of Neutrons by Helium and the $d-d$ Neutron Spectrum

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An investigation of the variation of the ratio of the scattering cross section of neutrons by helium and hydrogen has been made. Neutrons were obtained from a $d-d$ source partly surrounded by 3 cm of paraffin. The anomalously high cross section in helium at 1.0 Mev was found, and the helium-hydrogen cross-section ratio diminishes to about half this maximum value at 1.4 Mev. No other maxima were observed. The $d-d$ neutron spectrum was also investigated by helium recoils. The spectrum appears to be homogeneous; no evidence was found for a low energy group of neutrons with an intensity as great as 1 percent of the main group. This indicates that He^3 is not formed in an excited state.

IN an investigation of the α-particle spectrum obtained when Li^7 is bombarded by deuterons, Williams, Shepherd and Haxby[1] found that the continuous energy distribution had superposed on it a homogeneous group. The continuous spectrum was supposed to arise from the reaction ${}_3Li^7+{}_1D^2\rightarrow 2{}_2He^4+{}_0n'$, while the homogeneous group was ascribed to the reaction ${}_2Li^7+{}_1D^2\rightarrow{}_2He^4+{}_2He^5$. From the range of this group, the mass of the He^5 formed in the reaction is calculated to be 5.0137, which would make it unstable against disintegration[2] into an α-particle and a neutron by 0.84 Mev.

One may expect the scattering of neutrons by helium to be extremely high when the energy of the neutron is sufficient to produce an intermediate He^5 nucleus in the state formed in the Li^7 reaction; this should occur for a neutron energy of about one Mev, or five-fourths that of the energy of instability of He^5.

Staub and Stephens[3] have made a cloud-chamber investigation of this point by obtaining recoil tracks in helium and in ethane, using neutrons from the reaction ${}_4Be^9+{}_1D^2\rightarrow{}_5B^{10}+{}_0n'$. It is known[4] that this reaction yields four neutron groups, of maximum neutron energies 4.5, 4.0, 2.6, and 1.4 Mev at a bombarding energy of 0.9 Mev. By reducing the bombarding energy to 0.6 Mev, Staub and Stephens reduced the energy of the 1.4-Mev group to about 1.1 Mev at the point of maximum neutron intensity. This is very close to the neutron energy calculated to yield a resonance in the formation of He^5, and hence the ratio of the intensities of helium to proton recoils formed by this group might be expected to be large. This was found to be the case, and the ratio σ_{He}/σ_H of the backward scattering cross section of helium to hydrogen rose to about 9.5 at this point. With the bombarding energy increased to 0.88 Mev, the mean neutron energy of the group becomes 1.3 Mev, and the ratio of the cross sections at this point was observed to be about 6.5. At 2.3 Mev, their next measured point, the ratio dropped to its "normal" value of about 1.4, and remained nearly constant at this value up to about 6 Mev.

[1] J. H. Williams, W. G. Shepherd and R. O. Haxby, Phys. Rev. **52**, 390 (1937). Confirmed by H. Staub and W. E. Stephens, Phys. Rev. **54**, 236 (1938) and Phys. Rev. **54**, 237 (1938).

[2] A stable state of He^5 has been reported by F. Joliot and I. Zlotowski, Comptes rendus **206**, 1256 (1938), and J. de phys. et rad. **9**, 393 (1938).

[3] H. Staub and W. E. Stephens, Phys. Rev. **55**, 131 (1939).

[4] T. W. Bonner and G. Brubaker, Phys. Rev. **50**, 308 (1936).