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Radioactive Products from Gases Produced in Uranium Fission*

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Some radioactive products from the gases produced in the fission of uranium by slow neutrons have been studied as to their chemical identity and characteristic half-life periods. Two Rb activities have been observed, one of which decays with a period of 15.4±0.2 minutes into an active Sr with a half-life of 51±2 days; the other decays with a period of 17.8±0.2 minutes into an apparently inactive end product. The parent of the 15.4-minute Rb is a gas with a half-life of a few minutes whereas the other Rb arises from the decay of a 175±10-minute Kr. Absorption in Al indicates the maximum β-particle energy for these active products to be 3.8 Mev for the 15.4-minute Rb and 4.6 Mev for the 17.8-minute Rb. In a similar manner, a Cs has been observed which decays with a 32±0.5-minute period into a Ba which is inactive or of very long or short period with no evidence of the 300-hour product reported by Hahn and Strassmann. Data have been obtained which indicate that this long period Ba arises from the decay of a product of a very short period gas. This 32-minute Cs results from the decay of a Xe with a half-life of 17±1 minutes. Absorption in Al indicates a maximum β-particle energy of 2.6 Mev for this Cs activity.

INTRODUCTION

INVESTIGATIONS by Hahn and Strassmann and by Heyn, Aten and Bakker on the identification of the elements resulting from the slow neutron fission of uranium have resulted in the assignment of some of the observed radioactive periods to isotopes of rubidium and caesium. The latter were presumed to result from the β-disintegration of radioactive krypton and xenon. The present investigation was undertaken to determine the periods of these active gases and to establish genetic relationships between them and their products. In a preliminary report[1] it was indicated that the decay of a short period gas resulted in an active product with a half-life of about 18 minutes. This was attributed to the Rb of the same period reported by others.[2] Further work indicated, however, that an active Rb with approximately the same period could be obtained under conditions which pointed to a gas of much longer half-life as the parent substance. This report deals with the differentiation of these Rb activities and with a study of the 32-minute Cs previously reported.[2]

EXPERIMENTAL PROCEDURE

A 2.5-liter saturated aqueous solution of uranium nitrate, backed with paraffin, was irradiated with neutrons produced by protons on Be in the cyclotron. The radioactive gases resulting from the uranium fission were removed from the solution with a stream of air broken up into

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[1] G. N. Glasoe and J. Steigman, Phys. Rev. **55**, 982 (1939).

[2] O. Hahn and F. Strassmann, Naturwiss. **27**, 529 (1939); F. A. Heyn, A. H. Aten and C. J. Bakker, Nature **143**, 516 (1939).

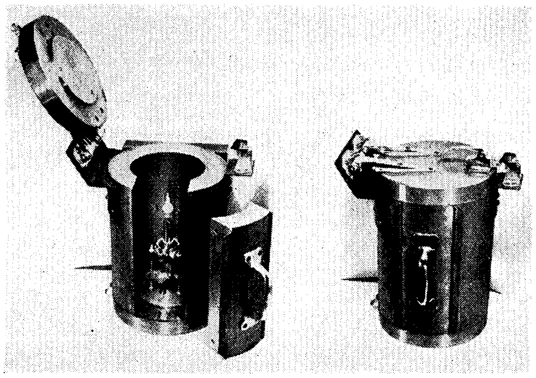

FIG. 1. Lead shield for Geiger-Müller counter.

small bubbles by means of a sintered glass disk or a perforated tube. In some instances the gases were collected after the irradiation. In other instances they were removed continuously during the neutron bombardment. This variation in procedure facilitated the differentiation of the products arising from the short period gases and those resulting from the decay of long period gases.

The products of the radioactive gases were concentrated and collected by a method first used by Rutherford[3] for obtaining the decay products of thoron. The procedure involves the collection of the positively charged recoil ions resulting from the disintegration of the active gas. To obtain the products of the long-lived gases the latter were collected in a metal vessel by water displacement and the recoil ions concentrated on a platinum wire made about 1000 volts negative with respect to the vessel. High activity of the products of the short period gases was obtained most satisfactorily by passing a continuous air stream through the solution, and into a metal cylinder containing the platinum wire, during the irradiation. Variation of the rate of the air flow provided a means of enhancing the activity of one product relative to others provided there was a sufficient difference between the periods of the active gases.

The chemical separation and identification of the decay products collected in the above manner was performed after the removal of the active deposit by immersing the platinum wire in hot water or some solution appropriate to the particular procedure. The chemical separations were carried out by following the standard method of adding inactive "carrier" salts of the elements in question. The active material was obtained either in the form of a precipitate or a residue resulting from the evaporation of a filtrate. After drying, the precipitate or residue was mounted on filter paper with cellophane tape and placed inside a brass tube which was then slipped over a Geiger-Müller counter. The counter tube[4] was made with a thin glass wall the inner surface of which was coated with chemically deposited silver to serve as the cathode. The GM counter was mounted in a thick lead shield as shown in Fig. 1 and connected to a scaling circuit with hard tubes, arranged so that scaling rates of 2, 4, 8, 16, or 32 could be selected depending on the activity of the material. Such a counter and associated circuit has proven to be extremely stable, with negligible sensitivity variations of long or short period, over many months. The support for the brass tube holding the active material was constructed so as to insure accurate reproducibility of the position of the active material

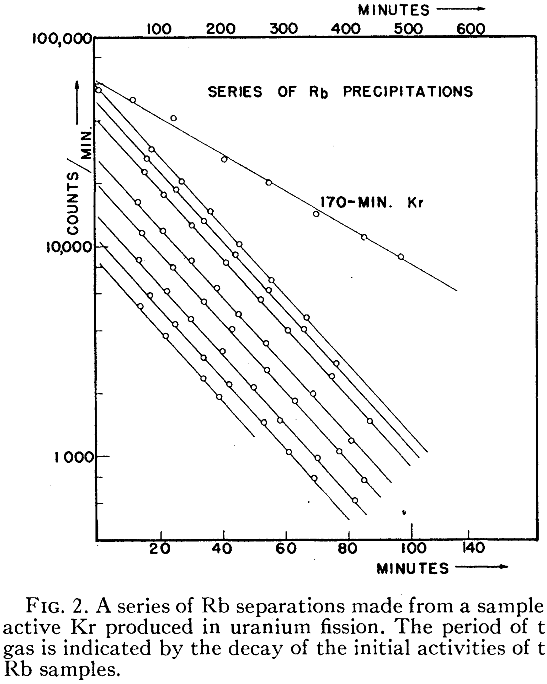

FIG. 2. A series of Rb separations made from a sample of active Kr produced in uranium fission. The period of the gas is indicated by the decay of the initial activities of the Rb samples.

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[3] E. Rutherford, Phil. Mag. 49, 161 (1900).

[4] The counter tube was made by Eck and Krebs, New York, New York. After pumping and baking the tube was filled with 8.5 cm of argon and 1 cm of alcohol.

relative to the counter tube. This precaution was particularly necessary in making absorption measurements.

Rb—17.8-Minute Half-Life

This isotope was one of the first products identified by Hahn and Strassmann,[2] and observed by Heyn, Aten and Bakker[2] to result from the decay of a gas produced in uranium fission. A Kr with a 3-hour period was observed by Langsdorf[5] and was assigned by Hahn and Strassmann as the parent of the Rb. The experiments described here were performed to determine this Kr period and to establish the genetic relationship between these two products. The procedure was to collect the active gas from the uranium solution a number of hours after the irradiation in order to eliminate the products arising from the shorter period gases. The waiting time was varied from about 3 hours to 12 hours with the result that the shorter time proved to be sufficient to obtain this active Rb without contamination, as determined by a comparison of the decay curves. The recoils from the disintegrating gas were collected for a definite length of time (15 minutes) on the platinum wire, and then washed off in a hot solution of saturated sodium bitartrate to which was added inactive RbCl. The rubidium bitartrate was precipitated by cooling the solution in ice. After filtering and drying the precipitate, the activity was followed for about four half-lives and extrapolated back to zero time (the time at which the wire was removed from the collection vessel). With the same sample of active gas the above procedure was repeated at 70-minute intervals, identical operations being performed in all respects. The series of decay curves obtained in this manner is shown in Fig. 2. The decay of the extrapolated initial activities in a number of such experiments indicates a half-life of 175±10 minutes, which is interpreted as the period of the Kr giving rise to this Rb activity.

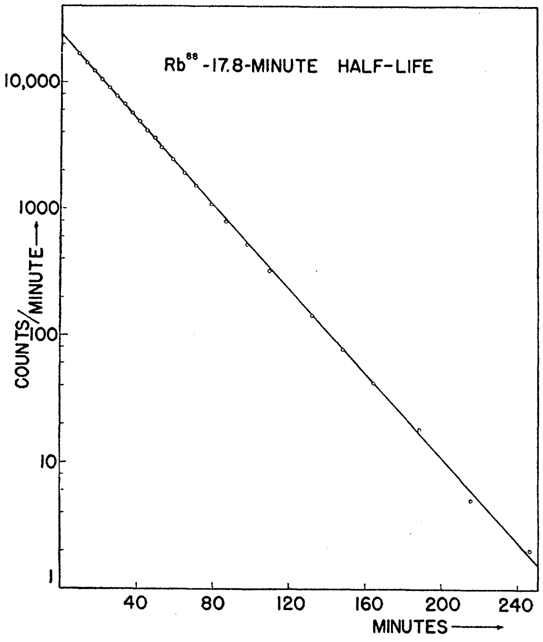

Fig. 3. Decay curve for the Rb product of the 175-minute Kr.

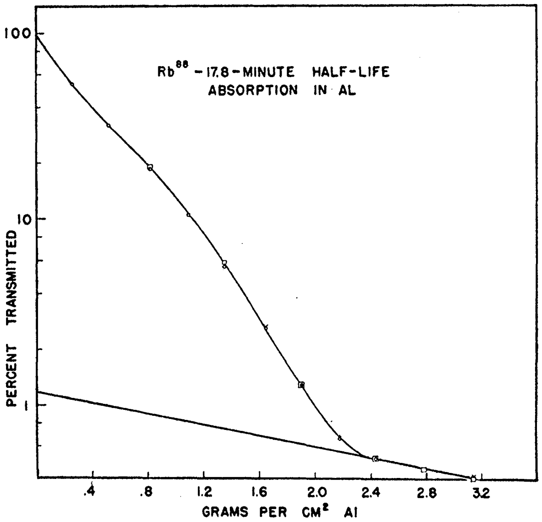

Fig. 4. Absorption in Al for the 17.8-minute Rb^{88}.

A number of experiments were performed to determine whether this Rb decays into an active Sr. A typical curve is shown, in Fig. 3, which is linear over 14 half-lives with the conclusion that the decay product is stable or that it has a period less than 5 minutes or greater than a year if the activity could be observed with the counter used. The half-life of this Rb isotope is found to be 17.8±0.2 minutes. This activity has been assigned to the isotope Rb^{88} by Hahn and Strassmann on the basis of their conclusion that the Sr product is stable.

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[5] A. Langsdorf, Jr., Phys. Rev. 56, 205 (1939).

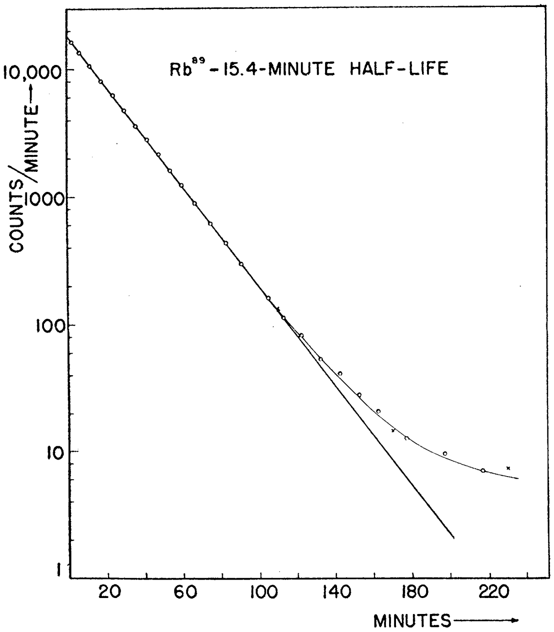

Fig. 5. Decay curve of Rb^{89} obtained from a short period Kr. The points marked with crosses were calculated for a 15.4-minute activity decaying into a 51-day product.

The absorption in aluminum of the β-particles from this activity was obtained by using cylindrical absorbers around the counter-tube with the active material mounted as indicated above under experimental procedure. An absorption curve is shown, in Fig. 4, in which the break is interpreted as giving the maximum range of the β-particles in Al. The range corrected for the absorption of the counter-tube wall is found to be 2.36 g/cm^2. According to Feather's[6] equation, $R=0.543E-0.160$, the maximum β-particle energy is 4.6 Mev. The fact that this energy appears somewhat high for such an activity might possibly suggest a very short period Sr as the decay product. The correction for the absorption of the wall of the counter was obtained by using a RaE source with the same absorbers and observing the shift of the absorption curve from that given by Feather.[6] This correction amounted to 0.06 g/cm^2.

Rb—15.4 Minute Half-Life

The high intensity of Rb activity resulting from a short irradiation (1 to 10 minutes) appeared inconsistent with that to be expected for the decay product of a long period gas. From absorption measurements it was apparent that there were two active Rb isotopes of approximately the same half-life. By applying the method of collecting the recoils during an irradiation, it was possible to obtain a Rb activity arising from a short period gas with inappreciable contamination due to the decay of the 3-hour Kr. This procedure, however, resulted in the complicating presence of intense activities due to Cs, Ba and Sr which had to be removed before the period of the Rb could be accurately determined. Because of the incompleteness of the separation of Cs from Rb by the known chemical methods (e.g. chloroantimonate, silicotungstate, etc.) it became necessary to adopt the following procedure; Rb was precipitated as the bitartrate in the presence of Cs, dissolved and reprecipitated five times, with additional inactive CsCl added each time. The fifth precipitate was dissolved in hot water to which was added inactive Ba and Sr chlorides. After two successive carbonate or sulphate precipitations to remove the Ba and Sr, the final filtrate was evaporated to dryness for the Rb activity. A typical decay curve for the material obtained in this manner is shown in Fig. 5 with the indicated half-life of 15.4±0.2 minutes. The active Sr into which this Rb decays was obtained under similar irradiation and collection conditions and was found to have a period of 51±2 days. It may

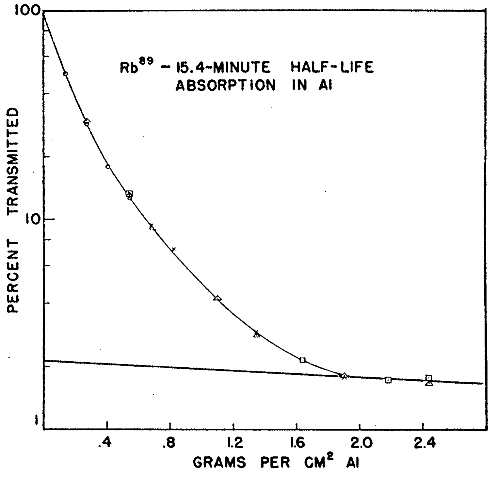

Fig. 6. Absorption in Al for the 15.4-minute Rb^{89} activity.

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[6] N. Feather, Proc. Camb. Phil. Soc. 34, 599 (1938).

be assumed that this is the same Sr isotope reported by Stewart[7] and by DuBridge and Marshall[8] as obtained from Sr(*d*, *p*) and assigned by them to Sr^{89}. The points marked with crosses in Fig. 5 were calculated on the basis of a 15.4-minute activity decaying into a 51-day product. From the manner in which this Rb activity is obtained it is estimated that the Kr parent must have a period of the order of one to five minutes.

An absorption curve for the β-particles from this Rb is shown in Fig. 6. The break in this curve indicates a corrected range in Al of 1.92 g/cm^2 corresponding to a maximum energy of 3.8 Mev. Due to the short period of this product and the loss of activity in the chemical procedure it was not possible to obtain a complete absorption curve on a single sample. The agreement obtained by using a number of samples of the Rb at different times is indicated by the multiple points in the case of several of the absorbers used. It might be remarked that in these absorption measurements several different sizes of the brass tube were employed to hold the active sample, with no marked effect being observed in the percent absorption due to the changed geometry between the counter and the sample. The counter tube was 2 cm in diameter and the brass tubes varied from 2.8 cm diameter to 4.8 cm diameter.

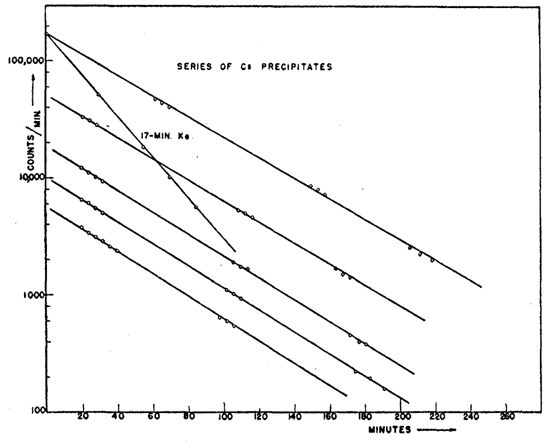

FIG. 7. A series of Cs separations made from a sample of active Xe produced in uranium fission. The period of the gas is indicated by the decay of the initial activities of the Cs samples.

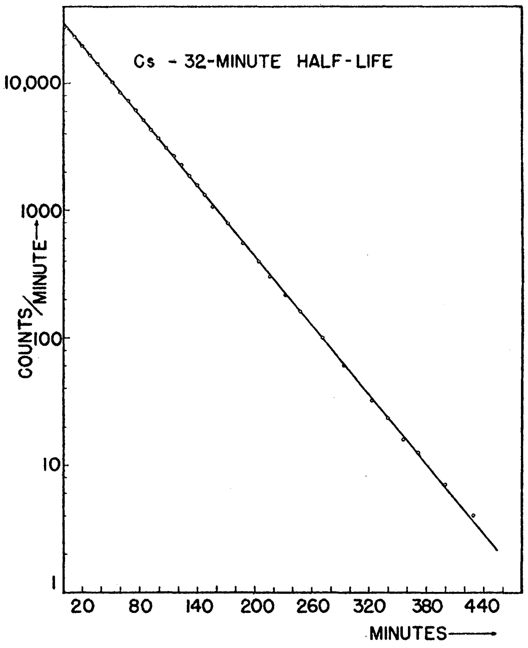

FIG. 8. Decay curve for the Cs product of the 17-minute Xe.

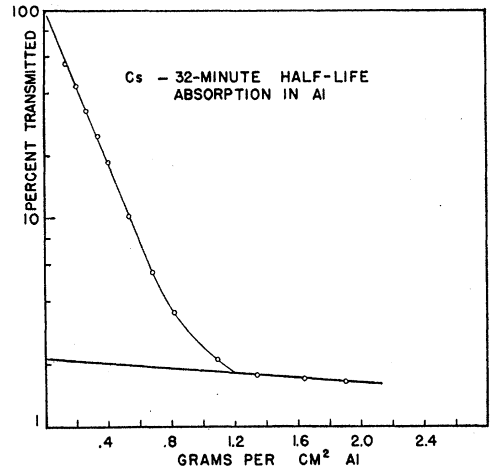

FIG. 9. Absorption in Al for the 32-minute Cs activity.

Cs—32-MINUTE HALF-LIFE

A Cs activity with a period of 30 to 33 minutes has been reported by Hahn and Strassmann and by Heyn, Aten and Bakker[2] and was studied in these experiments to determine the period of the parent gas. The procedure followed was in general the same as that employed to determine the half-life of the parent gas for the 17.8-minute Rb. In this case, however, the active gas was collected five minutes after the irradiation, in

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[7] D. W. Stewart, Phys. Rev. **56**, 629 (1939).

[8] L. A. DuBridge and J. Marshall, Phys. Rev. **56**, 706 (1939); **57**, 348 (1939).

order to avoid contamination by products from the very short period gases. The result is shown in Fig. 7. The extrapolated initial activities were plotted to obtain the decay curve of the parent Xe, showing a half-life of 17±1 minutes. Hahn and Strassmann estimated this period to be about 15 minutes from the rate of gas flow used in their experiments.

It was less difficult to obtain this Cs activity without contamination and of several chemical procedures tried the precipitation with silicotungstic acid from hot, 6*N* HCl solution was found the simplest and most rapid. A great many decay curves were taken for this activity with the result that there was no evidence for the formation of a 300 hour Ba as was reported by Hahn and Strassmann.[2,9] As shown in Fig. 8, the decay is linear over 14 half-lives and an activity ratio of 10,000. A long period Ba was observed, however, which decays into a La of about 40-hour half-life, by using the method of collecting recoils during an irradiation and with a very rapid air stream. This is probably the product reported by Hahn and Strassmann as arising from the 32-minute Cs. Measurements of the absorption in Al for this Ba activity indicated approximately the same absorption as observed for the Sr resulting from the decay of the 15.4-minute Rb. It is thus evident that this Ba activity should be observed by the departure of the Cs decay curve from linearity after about 9 half-lives. The conclusion to be drawn from these results is, therefore, that the 32-minute Cs decays into an inactive Ba or one whose period is less than 5 minutes or greater than a year.

An Al absorption curve for this Cs activity is shown in Fig. 9, with the range indicated to be 1.28 g/cm². This corresponds to a maximum energy for the β-particles of 2.6 Mev.

Conclusion

The results of these experiments may be summarized by the following genetic series for some of the radioactive products arising from gases produced in the slow neutron fission of uranium.

$$\mathrm{Kr}^{(88)}\xrightarrow{\text{3 hr.}}\mathrm{Rb}^{(88)}\xrightarrow[\text{4.6 Mev}]{\text{17.8 min.}}\mathrm{Sr}^{(88)}\ \text{probably stable,}$$

$$\mathrm{Kr}^{(89)}\xrightarrow{\text{1–5 min.}}\mathrm{Rb}^{(89)}\xrightarrow[\text{3.8 Mev}]{\text{15.4 min.}}\mathrm{Sr}^{(89)}\xrightarrow{\text{51 days}}\mathrm{Y}^{(89)}(?),$$

$$\mathrm{Xe}\xrightarrow{\text{17 min.}}\mathrm{Cs}\xrightarrow[\text{2.6 Mev}]{\text{32 min.}}\mathrm{Ba}\ \text{probably stable,}$$

$$\mathrm{Xe}\xrightarrow{\text{very short}}\mathrm{Cs}\xrightarrow{\text{very short}}\mathrm{Ba}\xrightarrow{\text{300 hr.}}\mathrm{La}\xrightarrow{\text{40 hr.}}\mathrm{Ce}(?).$$

Acknowledgments

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[9] O. Hahn and F. Strassmann, Naturwiss. **28**, 61 (1940).

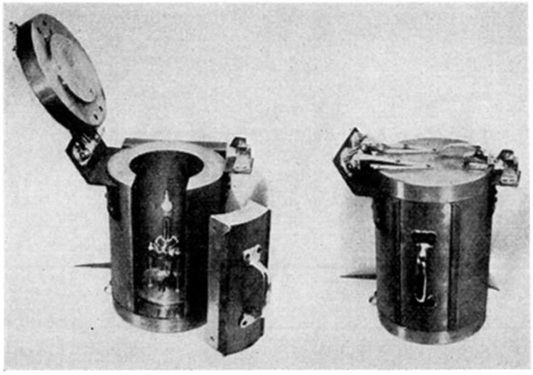

FIG. 1. Lead shield for Geiger-Müller counter.