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Determination of the Ranges of the Fission Fragments Emitting Delayed Neutrons. Chemical Identification of the 4.51-Sec. Delayed Neutron Activity*

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The ranges of the fission fragments leading to delayed neutron emission have been determined by absorbing the fragments in Al and counting the penetrating activity. The delayed neutron activities and their corresponding ranges in Al corrected for source thickness are: 4.51 sec., 4.05±0.03 mg Al/cm²; 55.6 sec., 3.98±0.06 mg Al/cm²; 1.52 sec., 3.63±0.12 mg Al/cm²; 22.0 sec., 3.21±0.04 mg Al/cm². Comparing the range of the 4.51-sec. activity with that of the 55.6-sec. Br, one finds that mass assignments possible for the 4.51-sec. activity are 86 to 90. Similarly, the range of the 1.52-sec. activity is compared to that of the 22.0-sec. I, and the possible mass assignments found are 129 to 135.

Chemical separation of Br from irradiated uranyl nitrate resulted in the identification of the 4.51-sec. activity as an isotope of Br.

1. INTRODUCTION

SNELL and co-workers[1] chemically isolated the delayed neutron activities of 55.6-sec. and 22.0-sec. half-lives in Br and I fractions, respectively, leaving three shorter-lived activities unidentified. Bohr and Wheeler[2] postulate that the neutron is emitted following the β^- emission of a fragment if the resultant nucleus is left in a state of excitation above the neutron-binding energy. The neutron emission occurs instantaneously from the excited nucleus, so that the period is determined by the half-life for β^- emission of the precursor. Thus, the 55.6-sec. Br decays to an excited state of Kr which then emits the neutron. Tentative mass assignments of 87 and 137 were given to the 55.6-sec. Br and 22.0-sec. I by Snell *et al.*[1] because of the possible identity of these activities with the 50-sec. Br^{87} and 30-sec. I^{137} found in uranium fission by Strassmann and Hahn.[3]

TABLE I. Half-lives and yields of the delayed neutron emitters.

Half-life	Neutron yield relative to fission neutrons (percent)
55.6 sec.	0.026
22.0 sec.	0.17
4.51 sec.	0.21
1.52 sec.	0.24
0.43 sec.	0.084

A summary of the data of Hughes and co-workers[4] on the half-lives and yields of the prominent delayed neutron emitters in the fission of U^{235} is given in Table I.

The chemical isolation of the delayed neutron emitters remaining to be identified is more difficult because of their relatively short half-lives. Since the task of devising chemical procedures which can be performed in from 10 to 30 seconds for isolation of the delayed neutron periods from among the 30 odd elements in fission appeared to be too laborious, it was decided to use another type of experiment to confine the neutron emitters among fewer elements.

The ranges of the fission fragments in Al have been determined for some of the masses formed in fission by the author and co-workers,[5] and it was found that the range varied with the mass, decreasing as the mass increased. Thus, the mass number of a fission product can be estimated by measuring the range and comparing it with ranges of fission products of known mass numbers. This technique was applied to the 4.51-sec. and 1.52-sec. delayed neutron emitters, with the

* This paper was first issued as an Argonne National Laboratory Report, CP-3622 (September 23, 1946).

[1] J. S. Levinger, E. P. Meiners, M. B. Sampson, A. H. Snell, and R. G. Wilkinson, Report CP-1967 (July 29, 1944); presented at the Chicago meeting, American Physical Society (June 20–22, 1946).

[2] N. Bohr and J. A. Wheeler, Phys. Rev. **56**, 426 (1939).

[3] F. Strassmann and O. Hahn, Naturwiss. **28**, 817 (1940).

[4] A. Cahn, J. Dabbs, and D. J. Hughes, Report CP-3094, (July 30, 1945).

[5] B. Finkle, E. J. Hoagland, S. Katcoff, and N. Sugarman, Report CC-2076 (August 25, 1944); Plutonium Project Record, **9B**, 6.6.2 (1946).

ranges of the 55.6-sec. and 22.0-sec. periods as standards. Once a rough mass assignment was found, it was used as a guide in selecting the chemistry to be used in isolating the activities. After the range of the 4.51-sec. period was found, it was shown that the activity could be separated chemically with the 55.6-sec. Br by extracting Br from irradiated uranyl nitrate solutions. The chemical isolation of the 1.52-sec. period was not attempted.

2. RANGE STUDIES

2.1 Experimental Procedure

The procedure used in the range studies is in outline as follows. A thin extended source of U, enriched in U^{235}, is covered with thin Al absorbers and a recoil catcher and is irradiated with thermal neutrons for a specified time. After irradiation, the catcher is separated from the source and absorbers, and the delayed neutron activity present on the catcher is determined. The delayed neutron curve is analyzed into its components, and the activity of each component is determined. The irradiations are carried out for varying thicknesses of Al absorbers (0, 1.70, 2.59, 3.40 mg Al/cm^2) so that the activity of each component as a function of Al absorption is determined. The technique used in these experiments differed somewhat from the range studies referred to earlier[5] in that the integral activity penetrating a given thickness of Al was determined directly here, whereas in the earlier experiments the Al absorbers themselves were analyzed, and the integral activity was obtained by summing over the individual absorbers. The irradiation times were varied from 5 seconds to 3 minutes for any given absorber in order to accentuate some periods relative to others. From the plot of activity *vs.* thickness of Al absorbers one finds the range of the fragments giving rise to a specific period, and from the range an estimate of the mass number. The thin absorber technique for determining ranges was adapted from that used by Segré and Wiegand[6] in their study of the maximum ranges of the fission fragments in various materials. A more detailed description of the apparatus follows:

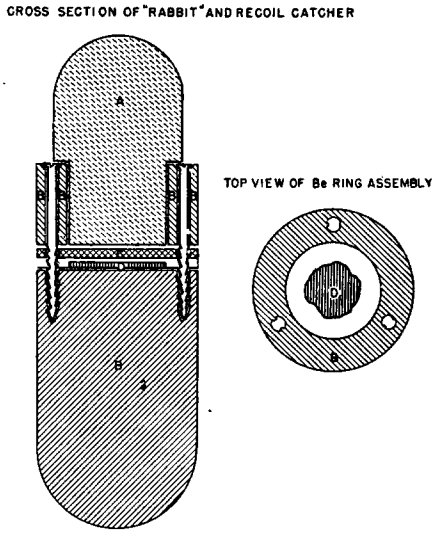

FIG. 1. "Rabbit" and recoil catcher. *A*, Bakelite plastic recoil catcher; *B*, beryllium; *C*, Al absorber; *D*, U^{235} source.

(a) *"Rabbit" and Recoil Catcher*

The short periods of the neutron emitters make it necessary to use means of introducing and removing the sample rapidly. The pneumatic tube at the heavy water pile of the Argonne Laboratory is well suited for this since samples can be blown in or out of the neutron field in about a second. The sample is contained in a "rabbit," the design of the one used in this experiment is given in Fig. 1. The "rabbit" was made of Be which is strong enough to endure the many shocks suffered in transit through the pneumatic tube. Since Be has a low activation cross section, it may be handled soon after removal from the pile. The source and Al absorbers are held in place by a beryllium ring which is attached to the "rabbit" by three screws. The recoil catcher made of fiber-impregnated Bakelite fits into the beryllium ring rather snugly so that it requires a jar to dislodge it. After irradiation, the "rabbit" is blown out of the pile and strikes a stop in the tube. Since the outer diameter of the recoil catcher is smaller than that of the beryllium "rabbit," the two separate and the former continues to the neutron counter. The activity on the catcher is

[6] E. Segré and C. Wiegand, Report LA-64 (February 29, 1944); recently published in Phys. Rev. **70**, 808 (1946).

TABLE II. Activity of the 55.6-sec. and 22.0-sec. periods on the recoil catcher as a function of Al absorber.

Period \ Al absorber (mg Al/cm²)	0	1.70	2.59	3.40	Range (mg Al/cm²)
55.6 sec.	8700 c/m	4690 c/m	3030 c/m	1120 c/m	3.93±0.06
22.0 sec.	50200 c/m	23900 c/m	8500 c/m	2000 c/m	3.16±0.04

then counted in the absence of the U^{235} source and Al absorbers.

The Al absorbers were 1.70, 2.59, and 3.40 mg Al/cm². The latter absorber was made up of two of the thinner ones. The individual absorbers were weighed, and the thickness used represents an average over the area confined by the "rabbit."

(b) *Neutron-Counting Apparatus*

The neutron counter used here was constructed by Hughes and Hall[7] for their study of the total yield of delayed neutrons from U^{235}. It consisted of a cadmium-covered paraffin block in which there were 8 boron-coated neutron counters arranged cylindrically around a sample slot. Tests with sources of high γ-activity showed that the counters were rather insensitive to γ-radiations. The scaler pulses activating the recorder could be varied by steps of 4, from a scale of 512 to a scale of 2. Since the decay of the samples soon after irradiation was rapid, the scaler pulses were recorded on a moving tape in an electro-cardiograph and later analyzed. After the sample had decayed for about a minute the scaler counts were recorded visually. The counter background was ~40 c/m with the pile operating at full power.

The bombardment and counting procedure is as follows. The "rabbit" is assembled with the source, Al absorber, and recoil catcher in place. It is put into the trap door of the pneumatic tube with the end of the "rabbit" opposite the recoil catcher facing the pile. The scaler is turned on and the electro-cardiograph started. The "in" button of the pneumatic tube is pressed, and the "rabbit" is sucked into the pile. Pressing the "in" button produces a pip on the electro-cardiograph tape. After the desired irradiation time the "out" button is pressed (also producing a pip on the tape which serves as the timing mark for the end of the irradiation), and the rabbit is blown out of the pile. A "stop" in the pneumatic tube, about 8 ft from the neutron counter, separates the "rabbit" from the recoil catcher which continues on toward the neutron counter and is stopped in the sample slot. The time elapsed between pressing the "out" button and the recording of the counting marks on the tape is about 1 second. As the counting rate decreases, the scaler output is varied from a scale of 512 to 8. After a minute has elapsed, the electro-cardiograph is turned off, but the scaler is kept on, and the counting is done by reading the recorder at specified intervals. The electro-cardiogram is developed and read, and the activity is plotted as a function of time. The curve is then ready for analysis into the component neutron activities.

2.2 Results and Discussion

The activities first studied by this technique were the 55.6-sec. Br^{87} and 22.0-sec. I^{137}, since their chemical identities were known and tentative mass assignments had been made. Irradiations were made for 3 minutes, and counting was started 20 seconds after the end of the irradiation. Six irradiations were made for each absorber, and the average decay curve was plotted. The analysis into the 55.6-sec. and 22.0-sec. periods was made by the technique used by Hughes *et al.*[4] of summing up the counts over a time interval (t_1 to t_2) where the 55.6-sec. period alone is present, and equating the sum to $\int_{t_1}^{t_2} A^0 e^{-\lambda t} dt$ from which the value of A^0 (activity at end of

TABLE III. Activity of the 4.51-sec. and 1.52-sec. periods on the recoil catcher as a function of Al absorber.

Period \ Al absorber (mg Al/cm²)	0	1.70	2.59	3.40	Range (mg Al/cm²)
4.51 sec. (15-sec. irradiation)	69000 c/m	41000 c/m	25000 c/m	9300 c/m	4.00±0.04
4.51 sec. (5-sec. irradiation)	38000 c/m	22000 c/m	13500 c/m	3800 c/m*	4.01±0.02
1.52 sec. (5-sec. irradiation)	53500 c/m	30000 c/m	14000 c/m	5000 c/m*	3.58±0.12

* These points were not used in the least-squares analysis because of the difficulty in obtaining reproducible results.

[7] D. J. Hughes and D. Hall, Report CF-3209 (October, 1945).

irradiation) can be calculated. After calculating the value of A^0 for the 55.6-sec. period one subtracts the activity of this period from the total activity and similarly analyzes the residual points for the value of A^0 for the 22.0-sec. period. The results given in Table II were obtained in this way.

The plot of activity *vs.* mg absorber for the recoil geometry used here (~50 percent) should be a straight line represented by the equation:

$$A_t = k(R-t),$$

where A_t is the recoil activity penetrating the absorber thickness t, k is a constant, and R is the range of the recoil fragments. The points establish fairly good straight lines (see Fig. 2), and the ranges found by a least-squares analysis of the points are 3.93±0.06 mg Al/cm² for the 55.6-sec. period and 3.16±0.04 mg Al/cm² for the 22.0-sec. period. The last point was not included in the analysis of the 22.0-sec. period, since it is beyond the range determined by the other points and arises from the straggling of the range of the fragments observed in earlier experiments.[5]

Shorter irradiations of 5 to 15 seconds were used for the 4.51-sec. and 1.52-sec. periods, and the activities of the 22.0-sec. and 55.6-sec. periods present in the sample were calculated from those found in the 3-minute irradiations. Here, again, between 6 and 10 irradiations were averaged for each decay curve. The 0.43-sec. period could not be isolated since the time interval before counting started was ~1 second, and it was difficult to reproduce such short bombardments. The decay curves were analyzed by the standard "peel-off" process using the half-lives given in Table I. The values of A^0 for the 4.51-sec. period for the different absorbers as found in the 15-second and 5-second irradiations, and those for the 1.52-sec. period as found in the 5-second irradiations are given in Table III along with the ranges determined by least-squares analysis.

TABLE IV. Corrected least-squares ranges of the fission fragments leading to delayed neutron emission.

Period	Range (mg Al/cm²)
4.51 sec.	4.05±0.03
55.6 sec.	3.98±0.06
1.52 sec.	3.63±0.12
22.0 sec.	3.21±0.04

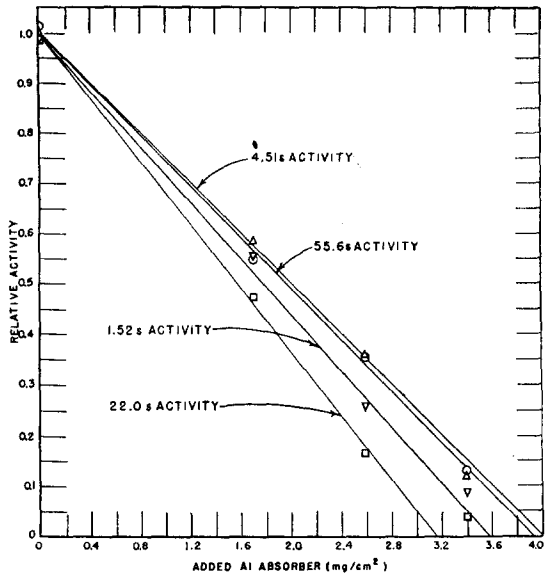

FIG. 2. Aluminum absorption curves of delayed neutron fission fragments.

○ 55.6-sec. period; least-squares range = 3.93±0.06 mgAl/cm².
□ 22.0-sec. period; least-squares range = 3.16±0.04 mgAl/cm².
△ 4.51-sec. period; (av. of 15-sec. and 5-sec. irradiations); least-squares range = 4.00±0.03 mgAl/cm².
▽ 1.52-sec. period; least-squares range = 3.58±0.12 mgAl/cm².

The data of recoil activity *vs.* mg Al absorber are plotted in Fig. 2. A correction resulting from the source thickness needs to be added to the ranges as determined in Al, namely one-half of the equivalent Al thickness of the source, or ~0.05 mg Al/cm². The corrected ranges are given in Table IV.

The ranges of mass numbers 87 and 137, tentatively assigned to the 55.6-sec. Br and 22.0-sec. I, as read off the smooth curve (range in Al *vs.* mass number) of the range experiment referred to earlier,[5] are 3.75 and 2.92 mg Al/cm², respectively. These ranges are about 7 percent lower than the ranges found for the fragments assigned to these mass numbers by the measurement of the delayed neutron activity. The discrepancy is probably due to error in the earlier experiments. In these, the Al absorbers were not weighed individually, but the average weight of the large sheets from which the absorbers were cut was used. Later, the variation in specific mass throughout a single sheet was found to exceed the variation in average weight among large sheets. The probable error in range thus

introduced was estimated at 10 percent. The earlier range results are more reliable on a relative basis, however, and can be used in a comparison of ranges from which the mass numbers may be obtained. A test of the relative comparison may be made by comparing the extremes in ranges of masses 87 and 137 with those found for the 55.6-sec. and 22.0-sec. periods. The ratio of the ranges of mass 87 to mass 137 from the smooth range curve is $3.75/2.92=1.28$. The error in this ratio is rather difficult to estimate but may be taken as about 1 percent. The ratio of the ranges of the 55.6-sec. and 22.0-sec. periods is $(3.98\pm0.06)/(3.21\pm0.04)=1.24\pm0.024$. These values agree as closely as can be expected since the differences in range compared are large, and the mass assignments used may be in error by several mass numbers. A comparison of more nearly similar ranges should yield even better agreement.

An approximate mass may be obtained for the 4.51-sec. period by comparing its range with that of the 55.6-sec. period. The dispersion of the ranges corresponding to the mass numbers in the light group is considerably smaller than in the heavy group; thus there is a larger error in the mass assignment in the light group. The range dispersion in the light group is 0.011 mg Al/cm^2/mass number, and in the heavy group, 0.054 mg Al/cm^2/mass number. The range of the 4.51-sec. period is 4.05 ± 0.03 mg Al/cm^2, or 0.07 mg Al/cm^2 larger than that of the 55.6-sec. period (3.98 mg Al/cm^2). The mass number of the 4.51-sec. period is therefore ~6 mass units smaller than that of the 55.6-sec. period, or 81 if a mass number of 87 is assigned to the 55.6-sec. period. The probable error in the mass number, arising from the probable errors in the ranges, is ±7 mass numbers. Many of the possible mass numbers of 81 ± 7 can be eliminated since the fission yield of the 4.51-sec. period of ~0.5 percent** exceeds the radiochemically determined yields of all masses up to mass 84.[8] Masses of 84 and 85 are also excluded, since their fission yields and/or those of the masses which would be formed by delayed neutron emission (83 and 84, respectively) are too small to allow for increments of the order of ~0.5 percent in their fission yields and still lie on the smooth yield *vs.* mass number curve found radiochemically. The remaining possible masses are 86 to 90, if we consider a possible error of 1 to 2 mass numbers in the mass assignment of the 55.6-sec. period. An inspection of the fission chains for these mass numbers indicates that the delayed neutron period should be separated chemically with Se or Br. The results of the chemical identification are given in Section 3.

The range of the 1.52-sec. period is 0.42 mg Al/cm^2 larger than that of the 22.0-sec. I. This corresponds to a difference of 8 mass numbers since the dispersion in the heavy group is ~0.054 mg Al/cm^2/mass number. The probable error in the mass number is ±4 mass numbers, somewhat higher than for the heavy group since the range curve has started to flatten in this mass range. Here, again, one may eliminate some of the possible masses in the range 129 ± 4 because of fission yield considerations. The remaining masses are 129 to 135 if again one considers a possible error of 1 to 2 mass numbers in the mass assignment of the 22.0-sec. period. Possible chemical elements with which the 1.52-sec. delayed neutron period may be separated are In, Sn, and Sb.

3. CHEMICAL ISOLATION OF THE 4.51-SEC DELAYED NEUTRON ACTIVITY

3.1 Se Isolation

Selenium carrier was added as $SeO_3^=$ to 3 g of UO_2Cl_2 in 5 ml of 4*M* HCl solution, and the mixture was irradiated for 5 seconds. After the irradiation, the active solution was mixed with 25 ml of 12*M* HCl, $NaHSO_3$ was added, and Se precipitated. The Se was centrifuged, and the precipitate counted in the neutron counter. The time elapsed between the end of the irradiation and the beginning of the counting was 45 to 50 seconds. The decay curve was that of an unseparated sample. Since the colloidal Se pre-

** For a discussion of the fission yields of the delayed neutron emitters see reference 8. The value of ~0.5 percent was arbitrarily adopted in this article for the fission yields of the 1.52-sec., 4.51-sec., and 22.0 sec. delayed neutron periods. The discussion would not be altered appreciably by fission yield values in the range 0.2 percent to 0.7 percent.

[8] Plutonium Project, "Nuclei formed in fission. Decay characteristics, fission yields, and chain relationships," issued by the J. Am. Chem. Soc. **68**, 2411 (1946); Rev. Mod. Phys. **18**, 513 (1946).

cipitate was removed from the solution by centrifugation and decantation, some of the solution stayed behind and was counted. An inspection of the gross decay curve of delayed neutrons (Fig. 3) shows that the decay of the 4.51-sec. activity would be obscured by the 22.0-sec. and 55.6-sec. activities after 45 to 50 seconds if 5 to 10 percent of the solution remained, even if the 4.51-sec. activity were quantitatively removed by the Se precipitation. Actually, the Se removal was far from complete, and an estimate of the residual solution was the order of 1 to 2 ml, enough to give the observed effect. Efforts to improve the decontamination and speed of the Se removal were not successful. Accordingly, a search for the 4.51-sec. activity in the Br fraction was undertaken.

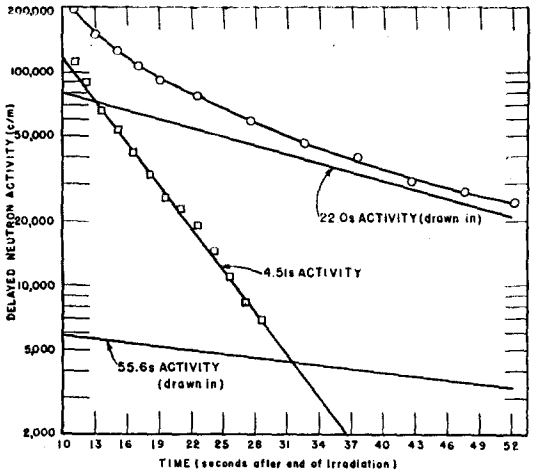

FIG. 3. Decay curve of delayed neutron activity of uranyl nitrate in 2 M Na_2CO_3 solution irradiated for 5 seconds.

○ Total delayed neutron activity.
□ 55.6-sec. and 22.0-sec. activities subtracted from smooth curve of total activity.

3.2 Br Isolation

In the isolation procedure the Br was extracted with CCl_4 after oxidation of I^- to IO_3^-. The method was adapted from that of Glendenin, Metcalf, and Novey[9] for the radiochemical isolation of I. A solution of uranyl nitrate in 2M Na_2CO_3, to which I^- and Br^- carriers are added, is irradiated for 5 seconds. After irradiation, the active solution is mixed with 10 ml of 2M Na_2CO_3 containing Br^- and BrO_3^- carrier and NaOCl in excess of that needed to oxidize the I^- to IO_3^-. The $Br^- - BrO_3^-$ mixture is stable in the basic solution. In separate experiments it was shown that I^- is quickly oxidized to IO_3^- by NaOCl in Na_2CO_3 solution and upon acidification, no I_2 is produced. Br^-, however, when present in the basic NaOCl solution is recoverable as Br_2 when the solution is acidified. Whereas the potential for oxidation of Br^- to BrO_3^- by NaOCl in basic solution is favorable, it was shown that the reaction does not occur during the time of this experiment. In addition, the BrO_3^- initially added as carrier remains as such throughout the procedure.

The solution is then added to a separatory funnel containing 50 ml of CCl_4 and 25 ml of 6N HNO_3. The solution becomes acid, Br_2 is liberated and is extracted in the CCl_4. Since it is present as IO_3^-, very little I is extracted. In the first experiments the solution in the separatory funnel was mixed by a stirring rod connected to a stirring motor, and the CCl_4 layer was siphoned over to a test tube placed in the sample slot of the neutron counter. In a series of six experiments, about 30 seconds elapsed between the end of the irradiation and the initiation of counting. Five to ten seconds of this time were consumed in the transfer of the solution from the separatory funnel to the counter. The radiochemical yields of Br, as determined by the ratio of the 55.6-sec. Br activity in the Br sample to that of an unseparated sample, averaged about 15 percent. Analysis of the neutron-decay curves showed that after 30-seconds decay the activity of the 4.51-sec. period was about that expected for a Br isotope when compared with the activity of the 55.6-sec. Br period, whereas the I activity was repressed by a factor of 2 to 6. The presence of I activity in the CCl_4 layer probably resulted from the suspension of aqueous solution because of the short time (5 to 10 seconds) allowed for the layers to separate. The chemical operations had to be completed in 30 seconds or less since by then the activity of the 4.51-second species had decayed roughly to that of the 55.6-sec. Br and was soon obscured by the latter and any 22.0-sec. I activity present as contaminant.

The Br separation procedure was modified in

[9] L. E. Glendenin, R. P. Metcalf, and T. B. Novey, Report CC-2218, (January 20, 1945); Plutonium Project Record **9B**, 8.21.3 (1946).

later experiments to achieve a higher radiochemical yield and more consistent results. Shaking of the separatory funnel by hand gave better extraction of the Br into the CCl_4 layer than mechanical stirring. Radiochemical yields of the Br samples counted varied from 15 to 42 percent and appeared to be determined by the fraction of the CCl_4 solution withdrawn for counting. The I activity was decontaminated by a factor of 4 with less variation than in the first Br separations. The results of these experiments are given in Table V.

TABLE V. Comparison of the activities of the delayed neutron periods in the Br fractions with a chemically untreated sample.

Activity after 30 seconds decay / Period	Activity (c/m) in Br fractions isolated from uranyl nitrate in Na_2CO_3 irradiated for 5 seconds	Average activity (c/m) from uranyl nitrate in Na_2CO_3 irradiated for 5 seconds	Radiochemical yield (ratio of column II to column III)
	No. 1: 3700		No. 1: 0.56
	No. 2: 1900		No. 2: 0.29
4.51 sec.	No. 3: 1700	6600	No. 3: 0.26
	No. 4: 2500		No. 4: 0.38
	av.*: 2771		av.*: 0.42
	No. 1: 2080		No. 1: 0.40
	No. 2: 1380		No. 2: 0.26
55.6 sec.	No. 3: 790	5250	No. 3: 0.15
	No. 4: 2200		No. 4: 0.42
	av.*: 1484		av.*: 0.28
	No. 1: 4200		No. 1: 0.087
	No. 2: 3450		No. 2: 0.071
22.0 sec.	No. 3: 1880	48400	No. 3: 0.039
	No. 4: 4800		No. 4: 0.099
	av.*: 3509		av.*: 0.072

* The average values were obtained from the arithmetic average decay curve of experiments 1–4 analyzed by a least-squares method.

The activities of the three delayed neutron periods in the Br fractions of experiments 1–4, given in Table V, were determined by a graphical analysis of the decay curves by the standard "peel-off" method. The average entry was determined by analyzing the average decay curve by a least-squares method, minimizing relative counting errors, since the time interval of counting was so varied during the decay that the number of counts in each interval was roughly the same. The results of the analysis of the average decay curve by the least-squares method are given in Fig. 4 for the early part of the decay curve where the least-squares curve of the 4.51-sec. activity is compared with the points determined by subtracting the sum of the 22.0-sec. and 55.6-sec. least-squares values from the smooth curve. The least-squares values of the 55.6-sec., 22.0-sec., and 4.51-sec. activities were determined from the complete decay curve extending to 6-minutes decay. The standard values in Table V used for comparison were determined from the average decay curve of an unseparated sample. A typical decay curve of a chemically untreated sample is given in Fig. 3 where the 55.6-sec. and 22.0-sec. activities were determined from the later portion of the decay curve extending to 8-minutes decay.

An examination of the data given in Table V shows that the radiochemical yield of the 22.0-sec. I activity is lowered appreciably relative to that of the 55.6-sec. Br activity as one would expect from the chemical operations performed. The yield of the 4.51-sec. activity, on the other hand, is roughly the same within experimental error as that of the 55.6-sec. Br activity; this demonstrates the chemical identity of the two isotopes. The fact that the average yield of the 4.51-sec. period is somewhat higher than that of the 55.6-sec. period might be indicative of a short-lived Se parent of the 55.6-sec. Br.

The mass assignment of the 4.51-sec. Br depends to some extent on that of the 55.6-sec. Br. In Section 2 it was shown that if the mass assignment of the 55.6-sec. Br is about 87, the mass of the 4.51-sec. period is in the range of 86 to 90. Mass 86 appears unlikely for a Br isotope since the calculation of the maximum β^- decay energy by the Bohr-Wheeler formula shows that the energy is about 1.5 Mev lower than the energy required to boil off a neutron from Kr^{86}. The remaining possible mass assignments of the 4.51-sec. Br are 87 to 90.

3.3 General Discussion

The delayed neutron emitters identified are divided evenly between the two groups of fission products, and their masses are such that they lie on the lighter ends of the mass groups.

The fission-yield *vs.* mass-number curve, determined by radiochemical analysis, appears to be a smooth continuous curve within the limits of experimental error of about 10 percent.[8] True discontinuities, although not observed, should exist because of the delayed neutron emitters. For example, the 4.51-sec. Br activity has a fission yield of ~0.5 percent; thus, the chain of the mass number of Br (87–90) should have its

yield lowered by ~0.5 percent which is about 10 percent of the fission yield of the chain in this region, and the chain of one mass number lower should have its fission yield raised by about 10 percent. Therefore, instead of a slowly rising fission yield at this mass, there should be a sharp discontinuity of about 20 percent. As yet no chemical evidence for this behavior has been found, but this technique may be applied to ascertain the mass numbers of the delayed neutron emitters more exactly. In this respect it should be mentioned that there is some uncertainty about the mass assignment of 137 to the 22.0-sec. I activity. The yield of Xe^{136} should be increased by the delayed neutron yield of ~0.5 percent if the assignment is correct. On the other hand, the relative yield determination, made by Thode and Graham,[10] of the stable noble gases formed in fission shows that the yield of Xe^{136} is about 15 percent lower than that of Xe^{134}, whereas the inverse should be true, first, because of the slow increase of yield with mass number in this region and, second, because of the contribution of the delayed neutron yield. The gas-yield results seem to indicate that the mass assignment of the 22.0-sec. I is 136. If this is true, then the Xe^{135} formed from the 22.0-sec. I should have an independent yield of ~0.5 percent. This is in rough agreement with the independent yield of ~0.3 percent for Xe^{135} determined by Hoagland and the author[11] by measuring the independent Xe^{135} activity directly.

[10] H. G. Thode and R. L. Graham, Report MX 129 (April, 1945); recently published in Can. J. Research **25**, 1 (1947).
[11] E. J. Hoagland and N. Sugarman, Report CC-3007, (April 15, 1945); Plutonium Project Record **9B**, 7.37.3 (1946).

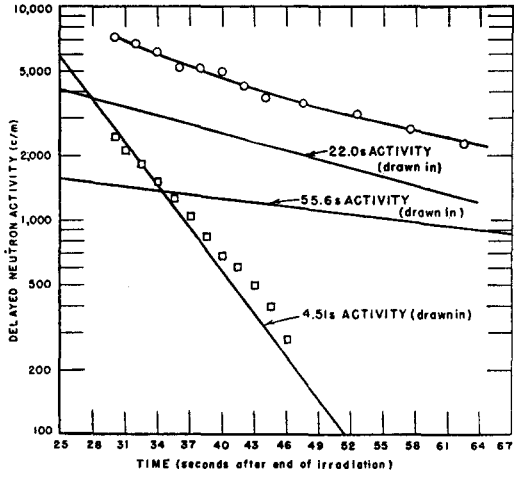

FIG. 4. Decay curve of delayed neutron activity of Br fraction isolated from uranyl nitrate in 2 M Na_2CO_3 solution irradiated for 5 seconds (least-squares analysis of average decay curve of 4 irradiations).

○ Total delayed neutron activity.
□ 55.6-sec. and 22.0-sec. activities subtracted from smooth curve of total activity.

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