Delayed Neutrons in Plutonium and Uranium Fission

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Samples of Pu^{239} and U, enriched in the 235-isotope, were irradiated in the graphite pile at Argonne. The decay of the delayed neutrons following fission was recorded on a magnetic wire recorder. Analysis of the decay data, 18 runs with Pu^{239}, and 37 with U, gave the following expression for the decay curves:

$$A_t = C_{Pu}(1.2e^{-0.660t} + 1.1e^{-0.154t} + 1.0e^{-0.0309t} + 0.10e^{-0.0127t}),$$

for Pu^{239}, and:

$$A_t = C_U(1.65e^{-0.613t} + 1.36e^{-0.156t} + 1.00e^{-0.0308t} + 0.15e^{-0.0126t}), \text{ for } U^{235},$$

when all the periods are irradiated to saturation.

Within the limits of experimental error, the half-lives for Pu^{239} and U^{235} are identical, namely:

(55.0 ± 0.4) sec., (22.5 ± 0.3) sec., (4.45 ± 0.15) sec., and (1.10 ± 0.06) sec.

The error limits given were determined from the internal consistency of the various values of the periods, not from the statistical fluctuations in the counting procedure.

The change in the yield of the 55.0-sec. period relative to that of the 22.45-sec. period, in going from U^{235} to Pu^{239}, is of the proper sign but small in magnitude as compared to the change predicted from the fission yields for the known mass numbers of these delayed-neutron emitters. A comparison of the relative number of delayed neutrons per fission gives the ratio of the number from Pu^{239} to the number from U^{235} as 0.5.

1. INTRODUCTION

THIS study of delayed neutrons emitted from fission fragments was undertaken to determine the decay curve associated with Pu^{239} and to verify the reported values for U^{235}.

Previous work[1] gave the following analysis for the delayed neutrons from U^{235}:

$$A_t = C_U(1.2e^{-0.28t} + 1.2e^{-0.099t} + 1.0e^{-0.029t} + 0.135e^{-0.012t}),$$

with the 57- and 24-sec. periods admittedly more accurate than the 7 and 2.5 sec. ones. Subsequent work[2] resulted in the following analysis, the result of a one-second irradiation to better resolve the shorter periods:

$$A_t = C_U(2.37e^{-1.38t} + 1.67e^{-0.154t} + 1.00e^{-0.030t} + 0.083e^{-0.014t}),$$

representing periods of 50, 23, 4.5, and 0.5 seconds.

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[1] A. H. Snell, A. V. Nedzel, and H. W. Ibser, C-81, May 16, 1942. *C*-numbers refer to reports in project files; unless otherwise indicated, they have not as yet been published elsewhere. Such references are included to indicate appropriate credit and chronology. See H. D. Smyth, Rev. Mod. Phys. **17**, 459 (1945).

[2] A. H. Snell, M. B. Sampson, and J. S. Levinger, CP-1014, Oct. 28, 1943.

2. EXPERIMENTAL PROCEDURE

a. The Apparatus

Figure 1 indicates the experimental arrangement near the pile. A BF_3 proportional counter was mounted in a paraffin block 18×24×24 inches. The source was placed in an 8-in. lead cube, in such a position that there were 4 in. of lead between the source and BF_3 counter. Also, between the lead and counter there were 2 in. of paraffin. The lead served to reduce the intense γ-ray background, the paraffin to thermalize the neutrons for more efficient counting. An Armour (G.E.) magnetic wire recorder was used in determining the decay curves. It afforded two conveniences. In recording the scaler pulses, it made a permanent recording which could be used immediately for reduction; this is a distinct advantage over the photographic method of recording. Also, the record could be replayed at half-speed; this increased the accuracy in measuring the time interval between pulses.

The sources used in this experiment were contained in air-tight aluminum cylinders, one inch in diameter and height. The sample of U was considerably enriched in the 235-isotope.

b. The Taking of Data

The samples were irradiated in the graphite pile, five feet from the inner edge of the concrete shield. For high intensity, long duration runs to determine the longer periods, the apparatus was set up about 40 yards from the pile and shielded from it by a number of concrete walls in order to reduce the background.

For measurement of the shorter periods, the apparatus was placed beside the pile as shown in Fig. 1. The samples were withdrawn from the pile by means of a falling-weight pulley system and guided through a bent aluminum pipe to their final position in the counting geometry. The bend in the aluminum guide pipe was necessary in order to shield the counting geometry from the intense radiation coming out of the hole used for irradiating the samples. Initially the sample was withdrawn in 1.0 sec. By allowing an initial free fall of the weight, this time was later reduced substantially, to 0.6 sec.

The scaler pulses, which usually operate a mechanical recorder, were recorded instead, on the magnetic wire apparatus. The sample was in counting position when the weight hit the floor, at which moment the scaler was started. In order to eliminate the neutron background and possible γ-ray effects, runs were taken with, and without, a cadmium shield covering the counter, and backgrounds for each case. The net curve was then the result of the subtractions.

[(Run without Cd) − (background without Cd)]
− [(Run with Cd) − (background with Cd)]

TABLE I. Longest period.

Date	Relative intensity	Irradiation time	Half-life
		U^{235}	
5/19/44	1.00	6 min.	57.5 sec.
5/20/44	2.00	6	54.0
5/20/44	1.00	6	55.4
5/20/44	1.00	6	55.0
5/22/44	2.00	6	54.0
5/27/44	2.94	3	55.0
		Weighted mean:	55.1 sec.
		Pu^{239}	
Date	Relative intensity	Irradiation time	Half-life
5/23/44	1.50	5	51.9
5/23/44	2.00	6	56.0
5/23/44	1.50	5	55.5
5/24/44	1.50	5	54.7
5/24/44	1.50	5	55.5
5/24/44	2.00	6	51.2
5/25/44	5.00	3	58.0
5/27/44	5.00	3	55.0
5/29/44	5.00	3	54.9
		Weighted mean:	54.9 sec.
		Deviation:	$d/n^{\frac{1}{2}}=0.4$ sec.

The cadmium correction was always less than 1.0 percent of the run without cadmium.

After the decay curves were recorded, the rate of decay was obtained by measurement of the time between successive recorded scaler pulses (or groups of pulses) with a 0.1-sec. stopwatch. The stopwatch was started with the first pulse, and the time noted for each succeeding pulse. Usually this replaying of the recorded data was made at half-speed to increase the accuracy of the timing procedure. In recording,

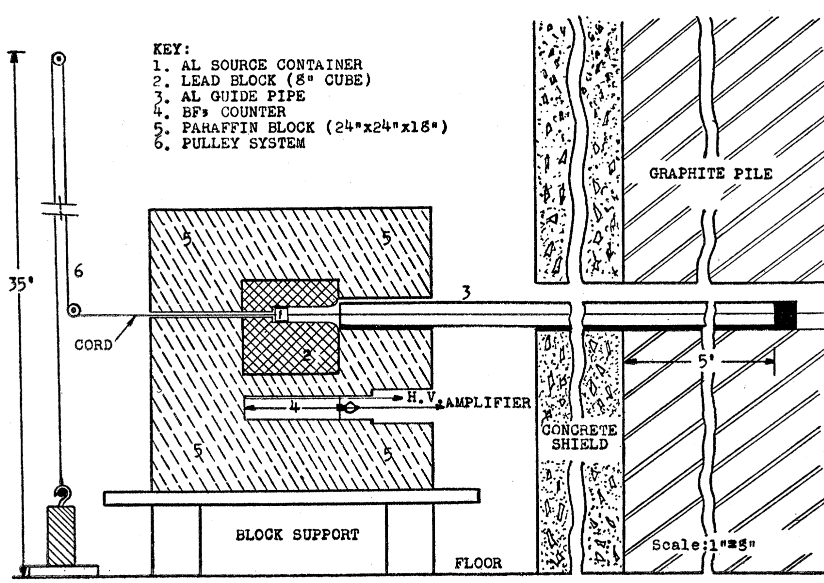

FIG. 1. Geometry of the apparatus for the measurement of the delayed neutrons (cross-sectional side view).

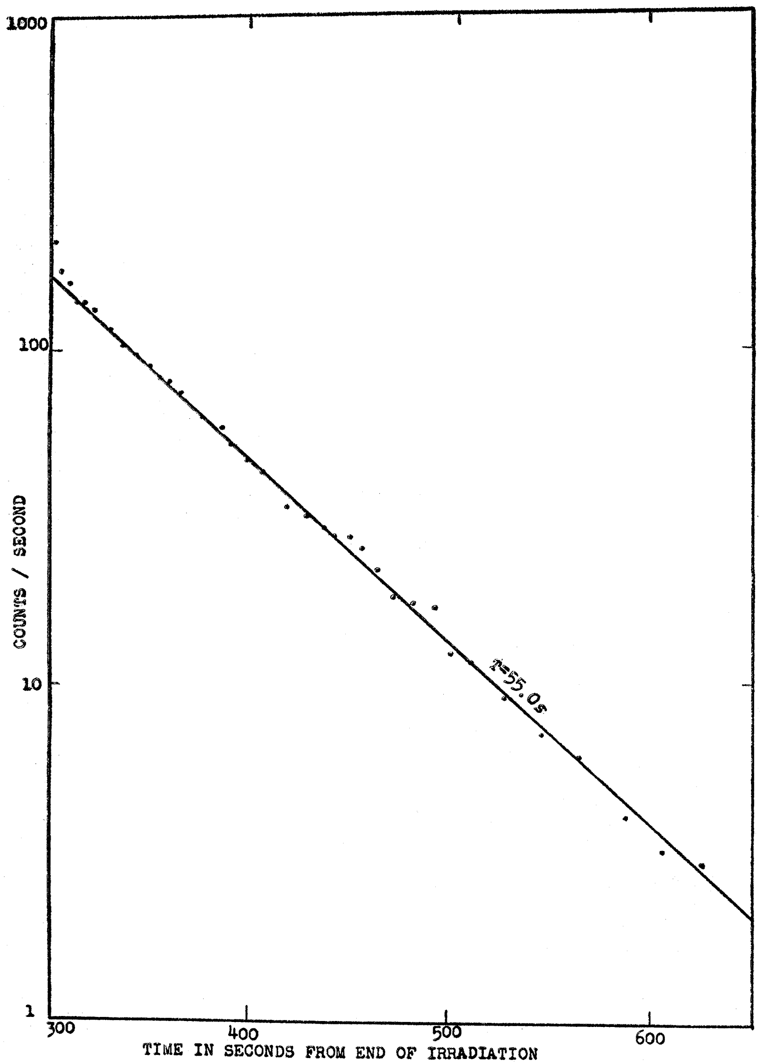

FIG. 2. Delayed neutrons from Pu^{239} (longest period only, with 3-min. irradation).

variable scaling was used to smooth out statistical fluctuations. Two independent reductions of each set of data were made, and any variation in results was re-examined.

3. DATA AND RESULTS

In these irradiations the intensity was varied by a factor of 1670 and the length of irradiation from two seconds to six minutes. The long and intense neutron bombardments were used in the study of the longer periods. For the longest period, counting was begun five to six minutes after removing the source from the pile in order that the contribution of the next longest period be less than 2 percent. (At $t=357$ sec. the contribution is 1 percent.) The data for one of these long period measurements are shown in Fig. 2, and the results for fifteen such runs are listed in Table I.

From these data the value of 55.0 sec. for the longest half-life was obtained and used in later reductions of the decay curves for determining the shorter periods. A second set of runs was made to determine the 22.5-sec. half-life. In these measurements the recorder was started 30 to 90 sec. after the end of irradiation. The medium intensity, shorter time irradiations were used to study the relative neutron yield of the various periods (Fig. 3 shows such a curve for Pu^{239}) and the least intense, shortest irradiations,

TABLE II. Half-lives in Pu^{239}.

Relative intensity	Irradiation time	T_1	T_2	T_3	T_4
2.5 (2 runs)	3 min.	55.0 sec.	22.2 sec.	3.9 sec.	1.07 sec.
2.5	30 sec.	55.0	20.6	4.4	1.16
2.5	13 sec.	55.0	24.3	4.5	1.03
1	1 min.	55.0	21.7	4.6	1.24
1	30 sec.	55.0	23.0	5.2	1.03
	Means:	55.0 sec.	22.5 sec.	4.5 sec.	1.10 sec.
	Deviations:	0.4 sec.	0.5 sec.	0.2 sec.	0.06 sec.

with the counting apparatus close by the pile, were used to best resolve the shortest periods.

Two methods were used in reducing the plotted decay curves. At first each run was reduced separately by the logarithmic "peeling off" process. Later, identical runs were made in groups of four or five, the curves plotted separately, and then the individual curves summed together. This composite curve was then used to determine the periods, again by the logarithmic "peeling off" procedure. The final results for the shorter periods are, for the most part, based on composite reductions. Such a reduction is shown in Fig. 4. The curve tailing off into a 22.4-second period is the sum of 9 individual curves after the 55.0-second half-life has been subtracted from each.

TABLE III. Saturated relative intensities for the half-lives in Pu^{239}

Relative intensities	Irradiation time	A_1	A_2	A_3	A_4
2.5	3 min.	0.101	1.00	0.955	0.78
2.5	3 min.	0.103	1.00	1.02	1.1
2.5	1 min.		1.00	1.07	1.1
2.5	30 sec.		1.00	1.06	1.6
1	1 min.		1.00	1.37	1.53
1	30 sec.		1.00	1.13	1.28
	Means:	0.10	1.00	1.09	1.2
	Deviations:	0.001		0.04	0.1
	Periods:	55.0 sec.	22.4 sec.	4.5 sec.	1.05 sec.

Because the Pu^{239} sample was available for only a short time, only nine runs were made on the short periods of Pu^{239}. Six of these runs were reduced individually, and the results are listed in Table II. The 55.0-sec. period was used with each. Next, the nine curves were added together and reduced compositely after applying to each the correction for the resolution time of the counting apparatus. The 55.0-sec. period was subtracted from each curve, and the curves were then added. From this composite curve the next period was determined and this value subtracted from the separate curves. The process was con-

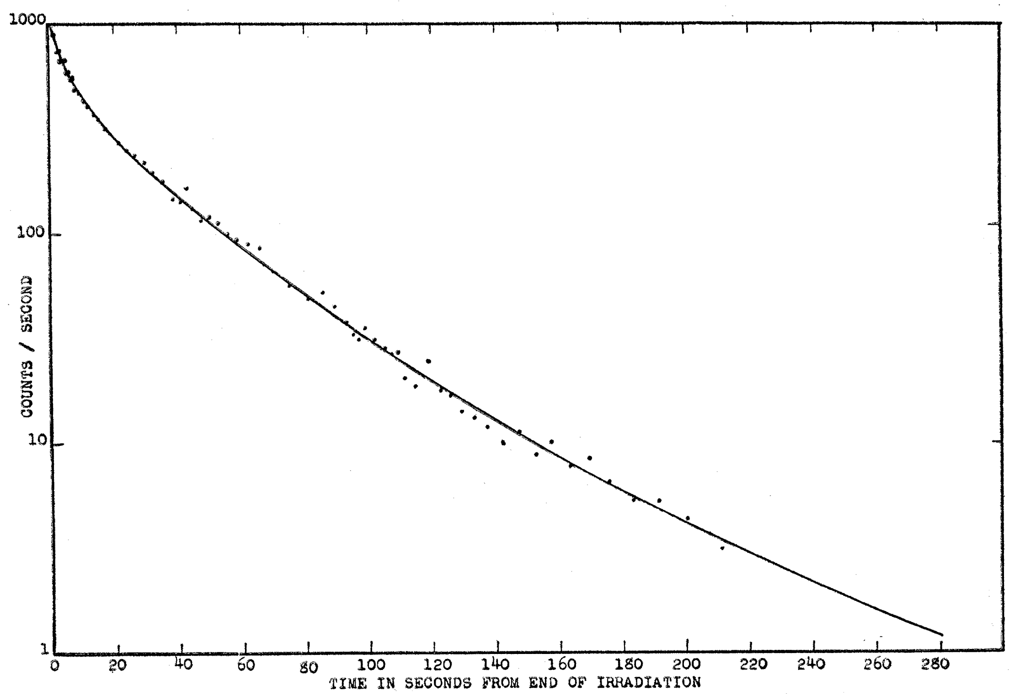

FIG. 3. Delayed neutrons from Pu^{239} (all periods present, 3-min. irradiation).

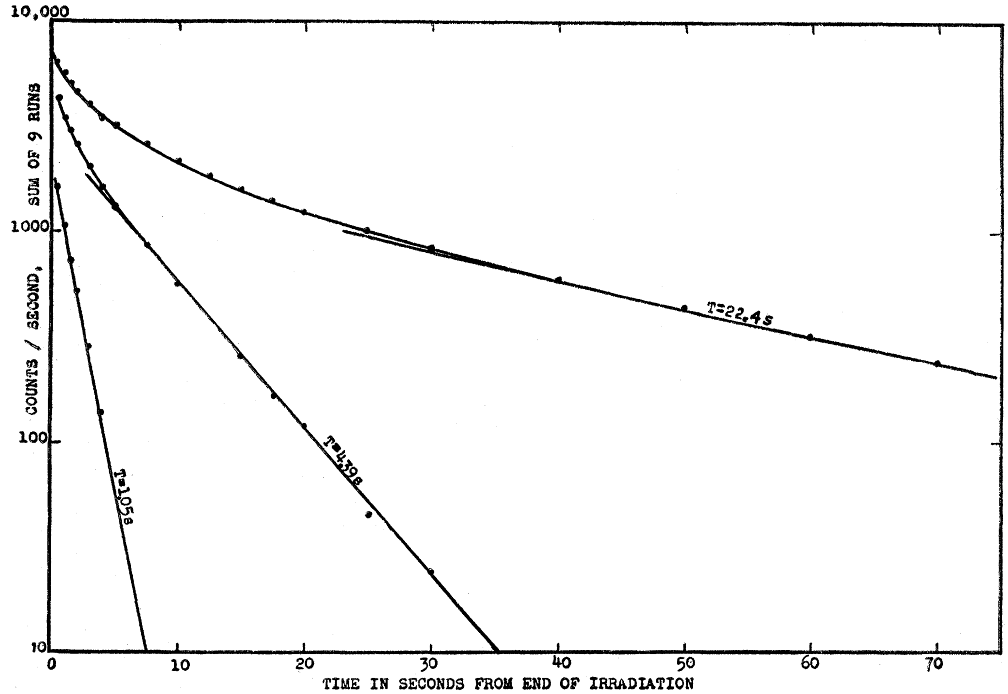

FIG. 4. Delayed neutrons from Pu^{239} (composite of 9 runs, showing the peeling-off procedure).

tinued until the final period was obtained. The resulting periods were:

55.0, 22.4, 4.6, and 1.04 sec.

A further reduction, subtracting periods from the composite curve only (no return to the separate curve to subtract each period) gave the following periods:

55.0, 22.4, 4.4, and 1.05 sec.

This last method of reduction should have introduced the least subjective error, since the data are treated a minimum number of times in this method.

The saturated relative intensities of the periods, A_i were found by extrapolating each period back to the end of the irradiation time and then computing the saturated values, using the previously determined half-lives. The results, relative to the value of $A_2=1.00$ for the 22.4-sec. period, are given in Table III. The final expression for Pu^{239} is:

$$A_t = C_{Pu}(1.2e^{-0.660t}+1.1e^{-0.154t}+1.0e^{-0.309}+0.10e^{-0.127t}).$$

Data on the shorter half-lives of U^{235} is more complete, since the U^{235} sample was available for a longer time. In Table IV are listed the values resulting from various reductions of 31 runs on U^{235}. For the most part, the various groups have been reduced compositely. The results for U^{235} are summarized in the expression:

$$A_t = C_U(1.65e^{-0.613t}+1.36e^{-0.156t}+1.00e^{-.0308t}+0.15e^{-0.0126t}).$$

The value of the constant C_U, the total delayed neutron yield for U^{235}, has been determined also.[3]

4. DISCUSSION OF RESULTS

In using the graphical method for reducing the gross decay curves, subjective errors are certainly introduced. For the values of the half-lives, as determined by the various methods for Pu^{239}, the agreement is very good. This internal consistency indicates that the composite method of reduction has adequately exploited the gross decay data. For a significant reduction of the

[3] D. E. Nagle, W. C. Redman, and D. Saxon, CP-2317, November 4, 1944.

errors, the statistics in the counting procedure itself must be improved.

One limitation of the experiment has been the resolving time of the counter system. This has been measured as 105 microseconds and corrected for, but the large magnitude of the corrections means that good resolution in the early part of the curve and statistical accuracy in the latter part were incompatible. Some of the Pu^{239} runs, in particular, have from 5 to 15 percent corrections in the early part of the curve; the early points on the U^{235} curves have corrections of the order of 5 percent. The apparatus was improved[4] and the resolving time decreased, but the Pu^{239} sample was not available for further measurements at that time.

Another shortcoming was the time required for removal of the irradiated source from the pile to the counting geometry. At first 1.0 seconds was required; later this value was reduced to 0.6 seconds. This removal time made detection of any half-life less than 0.5 seconds very difficult. It might be noted that recent reported values[5] substitute 1.8- and 0.44-second periods in place of the 1.10 second period. The longer periods are in good agreement with those reported here, but the removal time in this experiment prevented a confirmation of the shortest reported value.

These results indicate that the periods from both Pu^{239} and U^{235} are identical; hence, in all likelihood they are associated with the same fission fragments. A combination of the values of the half-lives for Pu^{239} and U^{235} gives:

$$(55.0\pm0.4)\text{ sec., }(22.5\pm0.3)\text{ sec., }(4.45\pm0.15)\text{ sec., and }(1.10\pm0.06)\text{ sec.}$$

The error limits given are those determined from the internal consistency of the various values of the periods, not from the statistical fluctuations in the counting procedure.

The chemical isolation of the two longest delayed-neutron periods,[6] together with the fission product yields of the associated mass numbers, leads one to expect a change in the delayed-neutron yield ratio of the two longest periods in going from U^{235} to Pu^{239}. The observed change in the yield of the 55.0-sec. period relative to that for the 22.5-sec. period has the proper sign but is smaller in magnitude than expected.

An approximate comparison of the relative number of delayed neutrons per fission for the two samples gives the ratio of the number from Pu^{239} to that from U^{235} as 0.5.

TABLE IV. Half-lives and saturated relative intensities for U^{235}.

T_1	A_1	T_2	A_2	T_3	A_3	T_4	A_4
6/13/44—4 runs, 3-min. irradiation							
55.0 sec.	0.153	23.8 sec.	1.00				
55.0	0.142	21.2	1.00	4.55 sec.	1.34	1.23 sec.	1.81
6/22/44—4 runs, 3-min. irradiation							
55.0	0.156	23.8	1.00	4.9	1.76		
55.0	0.146	21.2	1.00	4.43	1.33		
55.0	0.160	23.8	1.00				
6/23/44—5 runs, 3-min. irradiation							
55.0	0.171	23.6	1.00				
55.0	0.136	21.0	1.00	4.0	1.33		
55.0	0.120	22.0	1.00	5.35	1.42	0.92	2.36
6/29/44—4 runs, 3-min. irradiation							
55.0	0.133	21.5	1.00				
7/4/44—4 runs, 5-sec. irradiation							
		24.0	1.00	3.53	1.22	0.7	1.46
		22.4	1.00	4.0	1.38	1.08	0.95
		21.8	1.00	4.4	1.36	1.15	1.54
		22.0	1.00	4.1	1.38	0.97	1.57
		23.2	1.00	4.25	1.43	0.98	1.59
7/17/44—6 runs, 12-sec. irradiation							
		21.6	1.00	4.75	1.36	1.1	2.08
		23.0	1.00	4.7	1.34	1.5	1.47
		22.0	1.00	4.65	1.18	1.6	1.48
7/17/44—4 runs, 3-sec. irradiation							
		22.5	1.00	4.45	1.14	1.20	1.85
Means:							
55.0 sec.	0.146	22.47 sec.	1.00	4.43 sec.	1.36	1.13 sec.	1.65
Deviations:							
0.4 sec.	0.004	0.2 sec.		0.1 sec.	0.02	0.06 sec.	0.08

ACKNOWLEDGMENT

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[4] D. E. Nagle and D. Saxon, Monthly Report for October, 1944, CP-2301, November 4, 1944.

[5] M. T. Sampson and A. H. Snell, Monthly Report for July, 1944, CP-1934, August 3, 1944.

[6] A. H. Snell, J. S. Levinger, R. G. Wilkinson, E. P. Meiners, and M. B. Sampson, CP-1967, July 29, 1944. See Phys. Rev. **70**, 111 (1946).