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Short-Period Delayed Neutrons from Fission*

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A new group of delayed neutrons having a mean-life of 216±60 milliseconds has been observed to follow fission of U^{235}. The apparent yield is 2.7±0.7 percent of the delayed neutrons. The experiment was done using a multiplying assembly of uranium metal bombarded once every 30 seconds by x-rays from a betatron. A discussion is given of the hypothesis that these delayed neutrons follow the decay of Li^9, and that Li^9 is formed as the light fragment in ternary fission.

I. INTRODUCTION

THE five well-established delayed-neutron groups following uranium fission were accurately measured by Hughes, Dabbs, Cahn, and Hall.[1] Hughes *et al.* also reported the possible existence of a group of delayed neutrons having a mean-life (decay time to $1/e$ of initial value) of about 72 milliseconds. de Hoffmann, Feld, and Stein[2] did not observe this period, but did report a 9 millisecond period. Brolley, Cooper, Hall, Livingston, and Schlacks[3] found no gamma, beta, or neutron activities shorter than 0.62 second. Comments on these papers are made in Part VI, Discussion of Results.

The present work is an attempt to resolve the inconsistent results of previous investigators. Further incentive was provided by the availability of a new method of doing the experiment.

II. APPARATUS

The apparatus was arranged so that a burst of x-rays from a betatron produced neutrons in a highly multiplying assembly of U^{235} metal. In the assembly, fission chains sustained by prompt fission-neutrons produced delayed-neutron precursors during a time interval of less than 10 microseconds. A ten-channel time analyzer was used to study neutron leakage from the uranium assembly after the burst.

The betatron was pulsed every 30 seconds by a clock-actuated trigger. The x-ray yield was approximately 0.75 milliroentgen per burst at one meter, and the maximum x-ray energy was about 11 Mev. Length of the x-ray bursts was less than 1 microsecond. Distance from the x-ray source in the betatron to the center of the multiplying assembly was approximately 80 cm.

The multiplying assembly was a uranium sphere highly enriched in U^{235}. The uranium was not surrounded by reflecting material, except for its enclosure in a box of 0.030 inch cadmium. The cadmium box was a precaution against the introduction of a spurious period by the return of room scattered neutrons. Data were taken with the assembly adjusted to give a prompt neutron multiplication $M_p=100$. The mean life of neutron chains at this multiplication is less than 10 microseconds. Because of the hazard involved in operating an assembly at this high multiplication, the assembly was put together by remote control.

The neutron detector consisted of a block of paraffin 8 in.×13 in.×20 in., containing 26 General Electric proportional counters lined with B^{10}. Sensitivity to fission spectrum neutrons intercepted by the detector was about 10 percent; the corresponding sensitivity for counting gamma-ray photons from radium was about 10^{-7}. Because of the modest size of the paraffin moderator, and the large amount of B^{10} present in the counters, the mean life of neutrons in the paraffin block was less than 50 microseconds. The detector assembly was covered with 0.030-inch cadmium, and was located about 25 cm from the multiplying assembly, in a position where it did not intercept the x-ray beam.

Figure 1 is a schematic diagram showing the apparatus in the laboratory on the left, and the equipment in the control room on the right. The time analyzer was triggered by the betatron orbit-shift pulse, and there was a delay of one channel width between the trigger pulse and the opening of the first channel. The channel widths used to study delayed neutrons were 0.001, 0.010, 0.040, 0.100, and 1.000 seconds. All counts from the detector were recorded on a monitor scaler, which counted continuously. In addition, a gated "background" scaler counted for 1 second starting 26 seconds after the betatron was fired.

III. CORRECTIONS TO THE DATA

A background correction was applied to the data to take account of delayed neutron activity from the previous bursts; i.e., bursts preceding the opening of the channels by approximately 30 seconds, 60 seconds, 90 seconds, etc. The data from which the background correction was calculated were obtained from the background scaler. At 0.1 second after a burst, the correction amounted to 10 percent.

At the start and finish of each day, runs were made with 1-second channel widths. Other data taken the same day were normalized to the 1-second data taken

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* Work performed under the auspices of the U. S. Atomic Energy Commission.

[1] Hughes, Dabbs, Cahn, and Hall, Phys. Rev. **73**, 111 (1948).

[2] de Hoffmann, Feld, and Stein, Phys. Rev. **74**, 1330 (1948).

[3] Brolley, Cooper, Hall, Livingston, and Schlacks, Phys. Rev. **83**, 990 (1951).

that day by means of the number of monitor counts. This took care of variation in betatron yield during different runs.

Data taken with 0.001-second channel widths were tested to determine the time at which the counting apparatus was fully recovered from saturation during the neutron burst. This was done by setting a strong steady source close to the detector, which by itself gave a counting rate equal to that obtained from the multiplying assembly following a burst. Data were taken with this source in place, and the number of counts the source contributed was subtracted from each channel. It was found that the net counts per channel due to the multiplying assembly were the same within statistics with or without the strong steady source for all channels after 0.003 second. This indicates that the apparatus was fully recovered by the third channel.

One effect of neutron chains in the multiplying assembly is to cause larger fluctuations in the counting than would result from a random source. Analysis of repeated runs shows the points have a probable error about 4.5 times as large as expected from a random source. This is not the result of instability of the apparatus, which was checked with a random neutron source, but is inherent in the use of a multiplying assembly in the experiment. Table I lists the number of counts per point for the data shown in Figs. 2–4, and the "probable error" including the factor 4.5.

TABLE I. Probable error of experimental points.

Channel width (seconds)	Counts per point	"Probable error" of each point[a]	
		(percent)	(units)
1 (lowest points)	60 000	1.2	±0.5
0.1	32 000	1.7	±1.0
0.040	55 000	1.3	±0.8
0.010	4000	4.8	±4
0.001	2000	6.8	±6
Average of 0.001	16 000	2.4	±2

[a] Intercept on the ordinate on Figs. 2–4 is 86 units.

IV. MULTIPLYING ASSEMBLY

The use of a multiplying assembly was necessary in order to increase the yield of the experiment. The phenomena taking place in the assembly may be described as follows: during the x-ray burst from the betatron, prompt neutrons are created in the uranium by γ-n and γ-fission processes. This initial population of neutrons is sustained several microseconds by neutron chains, which create perhaps 200 times as many fissions as were produced by the betatron x-rays. A large number of delayed neutron precursors are formed during this "burst" at zero time. Delayed neutrons emitted after zero time initiate prompt neutron chains which, on the average, are 100 neutrons long, thus increasing the neutron leakage from the assembly by 100. The yield of the experiment is thus 200×100, or 20 000 times as large as would be obtained from a small mass of uranium in a nonmultiplying geometry.

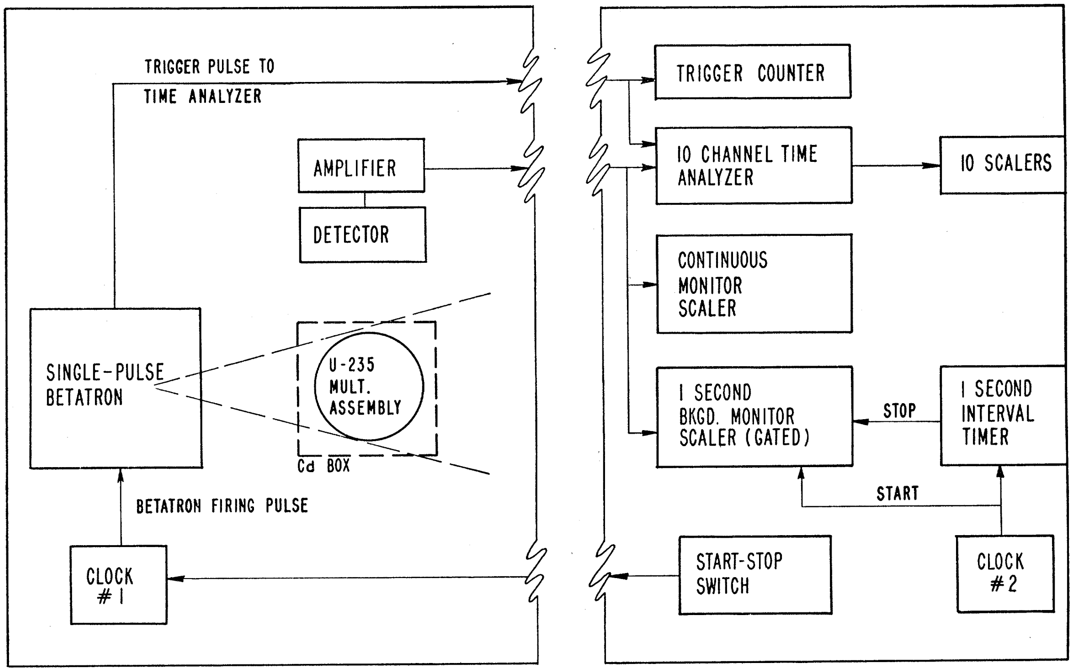

FIG. 1. Schematic diagram of apparatus. Equipment on the left was located in the laboratory, and equipment on the right was located in the control room.

TABLE II. Delayed-neutron groups and corresponding exponentials for a multiplying assembly. The A_j contain an arbitrary normalization factor. τ_i and β_i for the five longest delayed-neutron groups are from Hughes (reference 1).

		Multiplying-assembly exponentials ($M_p=100$)			
Delayed-neutron groups		Five delayed-neutron groups		Six delayed-neutron groups	
τ_i (sec)	$100\beta_i$	T_j (sec)	A_j	T_j (sec)	A_j
80.2	0.025	90.1	0.0141	90.1	0.0141
31.7	0.166	47.6	0.0472	47.6	0.0472
6.51	0.213	9.3	0.1543	9.3	0.1543
2.19	0.241	2.80	0.2098	2.80	0.2104
0.62	0.085	0.668	0.2476	0.669	0.2528
0.216	0.020			0.220	0.1802
Sums=	0.750[a]		0.6730[b]		0.8590[b]

[a] 100 times the apparent yield of delayed neutrons per fission neutron.
[b] These are the intercepts on the ordinate of the curves drawn in Figs. 2–5.

Inasmuch as the delayed neutrons are observed through the neutron chains they initiate, neutrons reaching the detector have the energy spectrum of leakage neutrons from the assembly, and not the energy spectrum of delayed neutrons. The apparent yield of one group of delayed neutrons compared with another group of delayed neutrons thus depends on the relative effectiveness of these groups of neutrons in initiating neutron chains in the uranium assembly. From measurements made with various neutron sources, it is known that the multiplication of the uranium assembly is constant within 5 percent for neutrons with energies from 100 kev up to the mean energy of the fission spectrum. The measured mean energies[1] of the established groups of delayed neutrons fall within this range. The energy spectrum of the new group of delayed neutrons reported here is completely unknown. If it falls generally within the range 100 to 1000 kev, then the apparent yield should not differ from the true yield by more than 5 percent due to energy sensitivity of the multiplying assembly. Energy sensitivity of the detector has no effect on the observed yield.

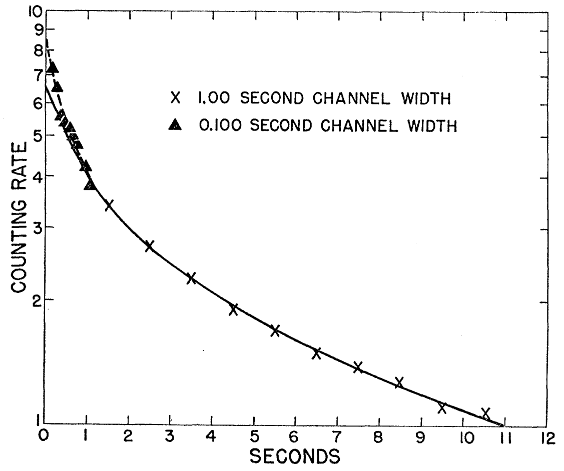

FIG. 2. Neutron leakage from the multiplying assembly followed out to 11 seconds.

Each group of delayed neutron precursors decays exponentially with a characteristic mean life, but the decay of the neutron population in a multiplying assembly is not proportional to the sum of these exponentials. After zero time, fission chains initiated by delayed neutrons create additional precursors. Thus, each delayed neutron period is coupled with all the other periods.

However, the decay of the neutron population in a multiplying assembly is proportional to a sum of exponentials,

$$n(t) \propto \sum_j A_j \exp[-t/T_j], \tag{1}$$

where $n(t)$ is the number of neutrons in the assembly, t is time, T_j are the periods, and A_j are the coefficients of the exponentials. The number of exponentials is the same as the number of delayed-neutron groups.[4] Letting τ_i and β_i represent the mean lives and yields of the delayed neutron precursors, the values of T_j are different from the τ_i, and the values of A_j are different from β_i/τ_i.[5]

The problem of transforming data taken from a multiplying assembly to the delayed neutron mean lives and yields (and *vice versa*) can be solved exactly by use of Laplace transforms.[6] The following equation ("inhour equation"), derived by means of Laplace transforms, was used to determine the values of T_j from assumed values of τ_i and β_i,

$$\sum_i \beta_i\tau_i/(T_j-\tau_i)=\sum_i \beta_i(1-k)/\delta k=0.003155. \tag{2}$$

k is the neutron reproduction constant, and δk is the change in k between delayed and prompt critical. For the coefficients of the exponentials,

$$A_j=2.383\frac{\sum_i \beta_i/(T_j-\tau_i)}{T_j\sum_i \beta_i\tau_i/(T_j-\tau_i)^2} \tag{3}$$

was used. The normalization factor 2.383 was arbitrarily chosen to make the decay curve of the multiplying assembly pass through 0.4 at 1 second when the values reported by Hughes[1] for τ_i and β_i are used. Decay periods of the multiplying assembly are compared with delayed neutron groups in Table II.

V. RESULTS

The experimental results are shown in Figs. 2–4. Where the statistical error is not indicated, the size of the symbol represents the error. The solid line in Fig. 2 is the predicted decay of the multiplying assembly calculated from the τ_i and β_i reported by Hughes[1] for five delayed-neutron groups having mean lives of 0.62 second and longer. The dashed line between zero and 1 second corresponds to six delayed-neutron groups.

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[4] The decay of the prompt-neutron population, several orders of magnitude faster, is not considered here.

[5] The convention that τ_i and T_j are positive numbers is used.

[6] G. Goertzel (unpublished); H. Soodak, *The Science and Engineering of Nuclear Power* (Addison-Wesley Press, Inc., Cambridge, 1949), Vol. 2, Chap. 8, p. 93.

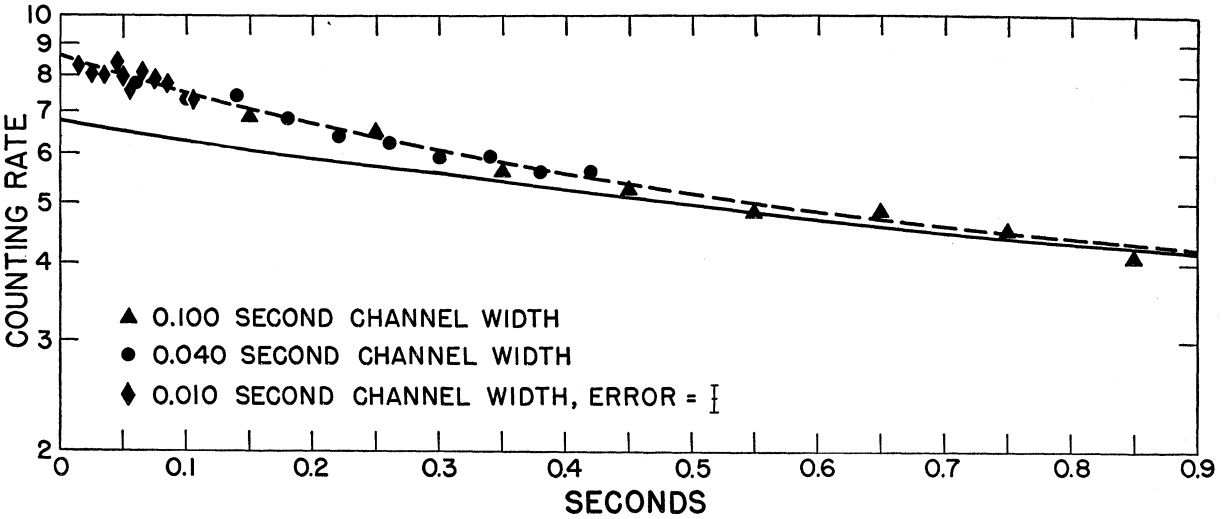

FIG. 3. Neutron leakage from the multiplying assembly.

Hughes measured τ_i and β_i for fission initiated by thermal-reactor neutrons, while the spectrum of neutrons causing fission in our multiplying assembly is approximately equivalent to a fission spectrum. Insufficient knowledge of the fission-product yield curve for fast-neutron induced fission of U^{235}, plus the fact that two of the five delayed-neutron emitters have not been identified, make it impractical to estimate the change (if any) in β_i between thermal-neutron and fission-neutron induced fission. However, the agreement after 1 second between experimental points and the solid line (Fig. 2) indicates the relative values of the five β_i measured by Hughes must be very nearly correct for our experiment.

The solid lines in Figs. 3 and 4 are also the predicted curve based on five groups. For times less than 1 second, the experimental points lie above this line. This indicates the existence of a sixth delayed-neutron group having a shorter mean life. The τ_6 and β_6 of the sixth group were determined by finding a predicted decay curve for the multiplying assembly which best fits the experimental points; this curve is shown by dashed lines on Figs. 2–4. In constructing this predicted curve, the Hughes values of τ_i and β_i for five groups were assumed to be exact, and only τ_6 and β_6 were adjusted to get the best fit.

It should be noted that T_j and A_j for the first five exponentials are somewhat different when calculated from six groups than when calculated from five groups. The sum of five exponential periods in the presence of six groups intercepts the zero time axis 1 percent higher than the solid line drawn in the figures. Therefore, the difference between the dashed line and the solid line is not exactly equal to $A_6 \exp(-t/T_6)$. The best fit to experimental points (Fig. 3) was obtained with $\tau_6=216$ milliseconds and $\beta_6=2\times10^{-4}=2.7$ percent of the total delayed neutron yield. Experimental uncertainties assigned to these values are ±60 milliseconds and ±0.7 percent. Values of τ_i, β_i, T_j, and A_j are tabulated in Table II.

The data taken with 0.001- and 0.010-second channel widths (Fig. 4) show that no delayed-neutron group was observed having a period between 5 and 50 milliseconds and an apparent yield as large as 0.5 percent of the delayed-neutron yield. The experimental results do not rule out the possibility that the sixth group reported here is actually a composite of two or more delayed-neutron groups having mean lives of approximately 200 milliseconds.

VI. DISCUSSION OF RESULTS

The experimental data reported by Hughes[1] as evidence for a delayed-neutron group having a mean life of 72 milliseconds have been plotted in Fig. 5, along with the same curves shown in Fig. 3. The different experimental procedure used by Hughes requires that the data be modified by four calculations before it can be compared with our results: (1) The Hughes five-period decay curve has been normalized to the same intercept on the zero-time axis as our five-period curve. (2) The Hughes five-period curve has been altered to conform with the decay curve for a multiplying assembly (Table II). (3) The displacement of the (renormalized) Hughes experimental points above the five-period decay curve has been multiplied by 1.504 to correct for decay of the sixth period during the 0.19 second irradiation of their uranium slug. The decay during irradiation was assumed to have a mean life of 216 milliseconds. (4) The Hughes experimental points have been raised (by less than 0.2 percent) because the sixth period in a multiplying assembly would decay at the rate of 220 milliseconds instead of 216 milliseconds. It is seen from Fig. 5 that the Hughes

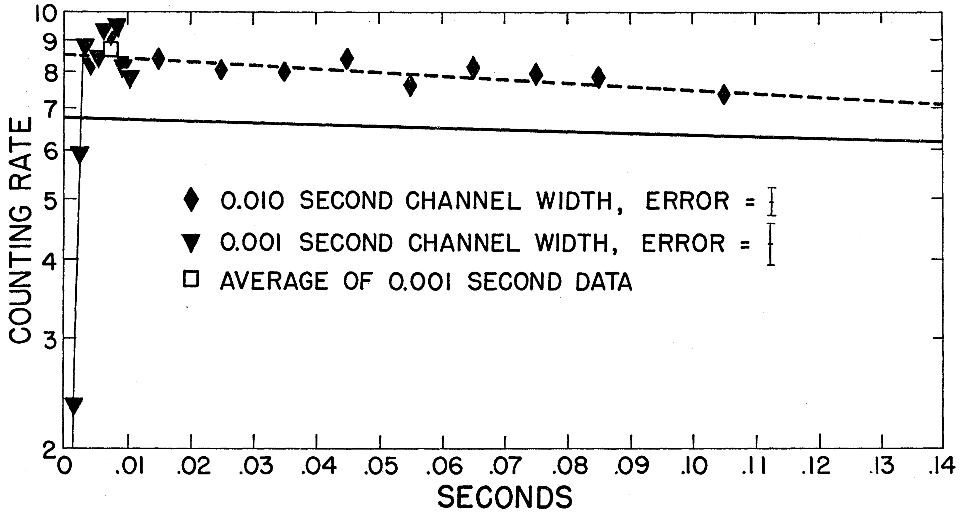

Fig. 4. Neutron leakage from the multiplying assembly. The first two points with 0.001 second channel width were taken before the apparatus was fully recovered from saturation during burst.

data are consistent with our results, except for the first point.

de Hoffmann[2] reported an activity with a mean life of 9 milliseconds and an apparent yield of 2 percent of the delayed neutrons. de Hoffmann states that the initial intensity of the short period after an infinitely short irradiation would be about 10 times that of the other periods together. Our data taken from 0.003 to 0.011 second with 0.001-second channel widths do not show such a period.

Brolley[3] looked for short period gamma- and beta-ray activities following fission, and also delayed neutrons by observing capture gamma rays from cadmium, with negative results. The efficiency of the paraffin block for returning thermalized neutrons to the cadmium converter may well have been too low for Brolley to detect a yield as small as observed here. The failure to observe a 200-millisecond gamma-ray period can be explained by hypothesizing that the delayed neutrons follow the decay of Li^9 (see below), in which case they presumably are not accompanied by the emission of a gamma ray. By using a different technique, Brolley might have observed Li^9 beta particles on those experimental runs in which the decay was followed from zero to 2.7 seconds. However, on these runs a 0.62-inch Al absorber was inserted between the uranium foil and the scintillator. Only a small fraction of Li^9 beta rays could penetrate this absorber.

VII. DECAY OF Li^9 AND TERNARY FISSION

According to beta-decay theory, it is unlikely for an isotope with mass greater than 70, such as products of binary fission, to have a 216-millisecond beta decay followed by neutron emission. The required high energy for so short a beta period would exceed the binding energy of a neutron and lead to emission of a prompt neutron. Neutron emission is more likely to follow short-period beta decay of low-Z isotopes.

It is hypothesized that the short-period delayed neutrons reported here follow the decay of Li^9, and that the Li^9 is formed as the light fragment in ternary fission. Li^9 is the only known delayed-neutron emitter having a mean life in the range 216 ± 60 milliseconds. Its mean life is 242 milliseconds.[7] The decay scheme is assumed to be

$$\mathrm{Li}^9\rightarrow\mathrm{Be}^{9*}+\beta,\quad \mathrm{Be}^{9*}\rightarrow\mathrm{Be}^8+n,\quad \mathrm{Be}^8\rightarrow 2\alpha.$$

Various schemes by which Li^9 could be formed from boron or other light elements in the counter tubes were considered, and all rejected on the basis that the threshold energies were too high for the reactions to be caused by fission neutrons or gamma rays, or by 11-Mev betatron x-rays. While mass numbers lower than about 70 are not observed for fragments of binary fission, Li^9 could be produced as the light fragment in ternary fission.

It is well established that a 10- to 25-Mev alpha particle is emitted in uranium fission with a frequency of about 1 event per 450 binary fissions.[8] There is some uncertainty regarding the occurrence in ternary fission of a light fragment having a mass and charge greater than an alpha particle. Titterton[8] reports observing three-particle fission in which the light fragment has a range less than 20 microns in photographic emulsion with a frequency of 1 event per 85 ± 10 binary fissions. The most probable value of the range is about 1 cm in air, and work with desensitized emulsions shows the tracks are still observed when the emulsion is no longer recording alpha particles. Allen and Dewan,[9] using counting techniques, report that in 1.3 percent of (slow neutron) fission events in U^{235} short-range charged particles considerably more massive than alpha particles are emitted. They conclude from the initial specific

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[7] Gardner, Knable, and Moyer, Phys. Rev. **83**, 1054 (1951); Holt, Thorn, and Waniek, Phys. Rev. **87**, 378 (1952); R. K. Sheline, Phys. Rev. **87**, 557 (1952).

[8] E. W. Titterton, Nature **168**, 590 (1951).

[9] K. W. Allen and J. T. Dewan, Phys. Rev. **82**, 527 (1951).

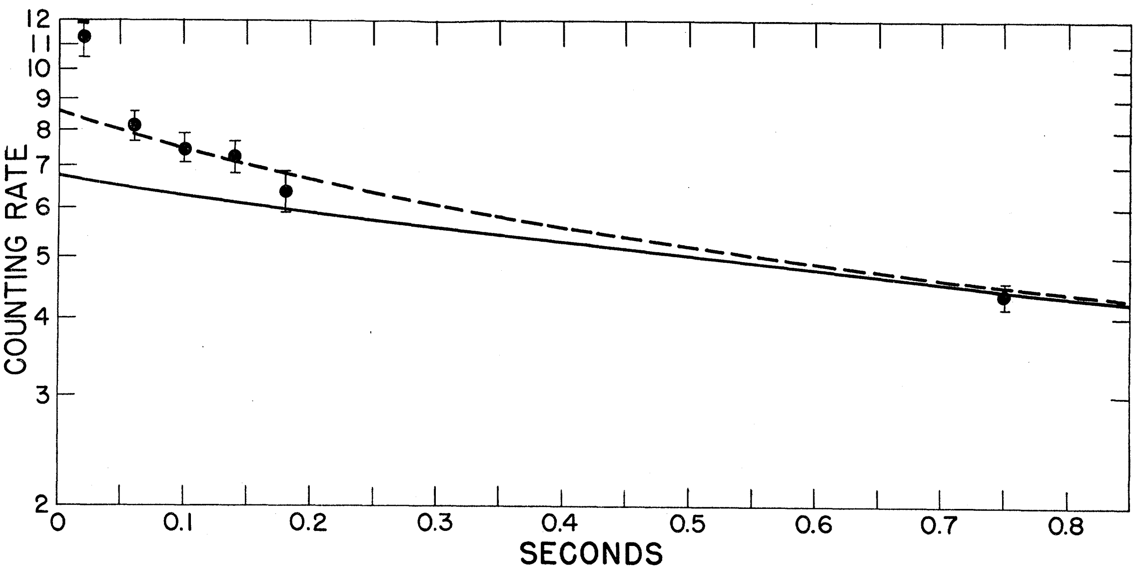

FIG. 5. Plot of the Hughes data (reference 1) with the curves shown in Fig. 3.

ionization of these particles that the mass may be in the range 13±4, but they add that photographic plate evidence is against the existence of particles with mass numbers greater than about 12. Laboulaye, Tzara, and Olkowsky,[10] using a cloud chamber, failed to observe any short-range tracks originating from the vicinity of the fission in excess of the expected nuclear recoils from the two heavy-fission fragments. Marshall[11] and Demers[12] have studied uranium fission extensively in photographic plates and have not reported evidence for this type of fission.

There are 20 known isotopes with masses between 5 and 12, and 24 isotopes in the range 13±4. If the evidence of Allen and Dewan is accepted, an order-of-magnitude estimate of the yield per fission of a particular light isotope such as Li^9 can be made by dividing 0.013 by 24, which gives approximately 5×10^{-4}. If one assumes the total neutron yield per fission is 2.5, this value is consistent with our observed yield of 2×10^{-4} for delayed neutrons with 216-millisecond mean life.

The fact that investigators of fission using photographic plates have not reported ternary fission with a hammer track has been presented as an argument against the formation of Li^9 in fission. The following is a possible explanation for their failure to see such tracks. If Be^{9*} is formed in the lowest excited level at 2.429 Mev,[13] then Be^8 will be formed in the ground state. The kinetic energy of Be^8 would be about 69 kev, since the neutron takes 8/9 of the kinetic energy released in the reaction $Be^{9*}\rightarrow Be^8+n$. The decay of Be^8 releases only 96 kev; the resulting two alpha particles would not make observable tracks in a photographic emulsion. This process for the decay of Li^9 leaves open the possibility that the frequency of observable hammer tracks in photographic emulsion is much lower than the frequency with which Li^9 is formed in ternary fission. A measurement of the energy spectrum of the Li^9 beta particles would help establish the likelihood of the process.

Further experiments looking for a delayed-beta period following fission, or for coincidences between delayed neutrons and beta or alpha particles, are necessary to substantiate or disprove the hypothesis that Li^9 is formed in ternary fission. A measurement of the energy spectrum of the delayed neutrons for comparison with the energy of neutrons from Li^9 decay would give supporting evidence.

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[10] Laboulaye, Tzara, and Olkowsky, Compt. rend. **237**, 155 (1953); J. de phys. radium **15**, 470 (1954).
[11] L. Marshall, Phys. Rev. **75**, 1339 (1949).
[12] P. Demers, Can. J. Phys. **31**, 78 (1953).
[13] F. Ajzenberg and T. Lauritsen, Revs. Modern Phys. **24**, 321 (1952).