Yield of Photo-Neutrons from U^{235} Fission Products in Heavy Water*

S. BERNSTEIN, W. M. PRESTON, G. WOLFE, AND R. E. SLATTERY
Clinton Laboratories, P. O. Box 1991, Knoxville, Tennessee

(Received January 20, 1947)

The photo-disintegration of the deuteron has been used to study the hard γ-rays emitted by fission products of U^{235}. The neutrons created in the process were used as the indicator of the presence of hard γ-rays. The fission products were placed at the center of a 10″ radius sphere of heavy water. Conclusions about the periods and yields of the hard γ-rays were made from the total number of photo-neutrons being captured in a large tank of oil surrounding the sphere of heavy water. Eight half-lives were found: 2.5 sec., 41 sec., 2.4 min., 7.7 min., 27 min., 1.6 hr., 4.4 hr., and 53 hr. The shortest one and longest one of these are least reliable. Eighty-five percent of the photo-neutrons appear in the two shortest half-lives, the 2.5-sec. component being three times as intense as the 41 sec. component. The total saturated activity of the photo-neutrons for an infinite amount of heavy water was calculated from the 10″ radius sphere measurements to be about 16.5 percent of the saturated delayed neutron activity. It is calculated that there must be of the order of one to two photons of energy above 2.2 Mev emitted per fission by fission products with half-lives greater than one second.

I. INTRODUCTION

THE threshold property of the photo-disintegration of the deuteron can be used as a crude γ-ray spectrometer. The reaction

$$\gamma + D \rightarrow n + p$$

requires that the energy of the γ-ray be at least 2.17 Mev[1] (the binding energy of the deuteron). The use of this reaction immediately "resolves" γ-rays above the threshold from those below, independently of the relative intensities of the two groups. It is convenient to use the neutrons created in the process as an indicator of the presence of hard γ-rays, since they may be slowed down by a moderating medium to thermal energies where they are easily detected. The total relative number of photo-neutrons produced is also a fairly easily determined quantity. It is a measure of the relative intensity of the hard γ-rays. We have studied the periods and yields of photo-neutrons produced in heavy water by the γ-rays from U^{235} fission products and from these yields have estimated the corresponding yields of the high energy γ-rays themselves.

II. APPARATUS

A schematic diagram of the apparatus is shown in Fig. 1. It consisted of a ten-inch radius, $\frac{1}{8}$-inch wall, aluminum sphere immersed in a large tank of oil. A "rabbit" containing a sample of enriched uranium oxide could be transferred from the center of the Clinton Laboratories chain-reacting pile[2] to the center of the sphere in about 0.25 sec. by means of a pneumatic tube. The sphere could be filled or emptied of heavy

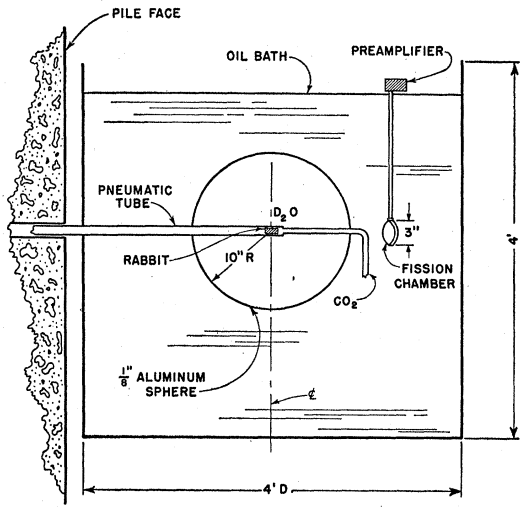

FIG. 1. Schematic diagram of apparatus.

* This document is based on work performed at Clinton Laboratory operated by Monsanto Chemical Company under contract No. W-35-058-eng-71 for the Manhattan Project and the information covered therein will appear in Division IV of the Manhattan Project Technical Series, as part of the contribution of Clinton Laboratories.

[1] F. T. Rogers, and M. M. Rogers, Phys. Rev. 55, 263 (1939).

[2] H. D. Smyth, *Atomic Energy for Military Purposes* (Princeton University Press, Princeton, 1945), Chapter VI; or Rev. Mod. Phys. 17, 392 (1945).

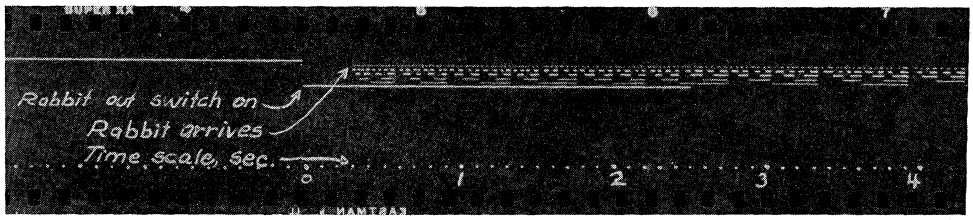

FIG. 2. Sample of film recorded data.

water in about one minute by means of a siphon arrangement. The number of photo-neutrons created in the sphere of heavy water by the fission product γ-rays was determined by studying the neutron distribution in the oil by means of fission chamber detectors. The chamber could be placed at chosen reproducible radial positions in the tank by means of a track and carriage arrangement. Since some of the periods involved are fractions of a second, it was necessary to keep the initial counting rates quite high in order to obtain good statistics throughout the run. An available amplifier and scaling circuit found to have a resolving time of 20 microseconds was adapted for use in the experiment. Exposure times and sample sizes were adjusted so that corrections for missed counts reached 5 percent only at the start of one or two of the runs. Fission pulses from the amplifier were fed into a scale of 256. The data were recorded by photographing the scaling circuit interpolation bulbs with an open shutter oscilloscope camera. The film moved past the neon bulbs in such a manner that each bulb appeared on the film as a dashed line for the period the bulb was lit. A time signal was impressed on the film every 0.1 second by means of a neon bulb and a revolving shutter driven by a synchronous motor. Another signal lamp connected through the electropneumatic control switch recorded the length of an exposure. Figure 2 shows a reproduction of a typical film.

III. METHOD

The method employed to measure the source activity is based on the following theory. Consider a point source of Q neutrons/sec., having an arbitrary energy distribution (above thermal energy), surrounded by an oil bath large enough to absorb all the neutrons. It is known that the absorption in the oil is caused almost entirely by capture of thermal neutrons by the protons. The number of captures of neutrons of energies higher than thermal is negligible. In the steady state the rate of production of neutrons must equal the rate at which they are captured, or $Q=\int(nv)_t\sigma_c dv$, where $(nv)_t$ is the flux of thermal neutrons at any point in the oil, σ_c is the macroscopic cross section for capture at thermal energy, and the integral is extended over the oil volume. Measurements with a fission chamber detector give a counting rate Y at any point in the oil which is proportional to $(nv)_t$, provided correction is made for the epicadmium counting rate.

For spherical symmetry, $Q\propto\int YR^2dR$. In our case, since there is negligible neutron absorption in the heavy water sphere, the integral is taken from R_0 to ∞, where R_0 is the sphere radius.

The ion chamber was fixed in a given radial position, the sample was sent into the pile for a fixed exposure time, then sent to the center of the sphere, and the resulting neutron counting rate recorded on the moving film as a function of time "t" after the end of the exposure. When the activity had decreased to an acceptable value, the ion chamber was moved to the next radial position and another exposure made with the same sample. A single series usually comprised exposures in four to eight positions. A complete experiment required three complete series: (a) with heavy water in the sphere, in which case both delayed and photo-neutrons are present; (b) with the sphere empty, so that the activity is that caused by delayed neutrons alone; and (c) with the sphere empty and the fission chamber surrounded by cadmium, in order to measure the delayed neutrons with epicadmium energies. With heavy water in the sphere the number of epicadmium neutrons was negligible.

The constancy of the pile power level and the detecting instruments was checked by repeating the initial exposure at the end of each experiment. Instrument sensitivity alone was checked by means of a Ra-α-Be source. Variations in the

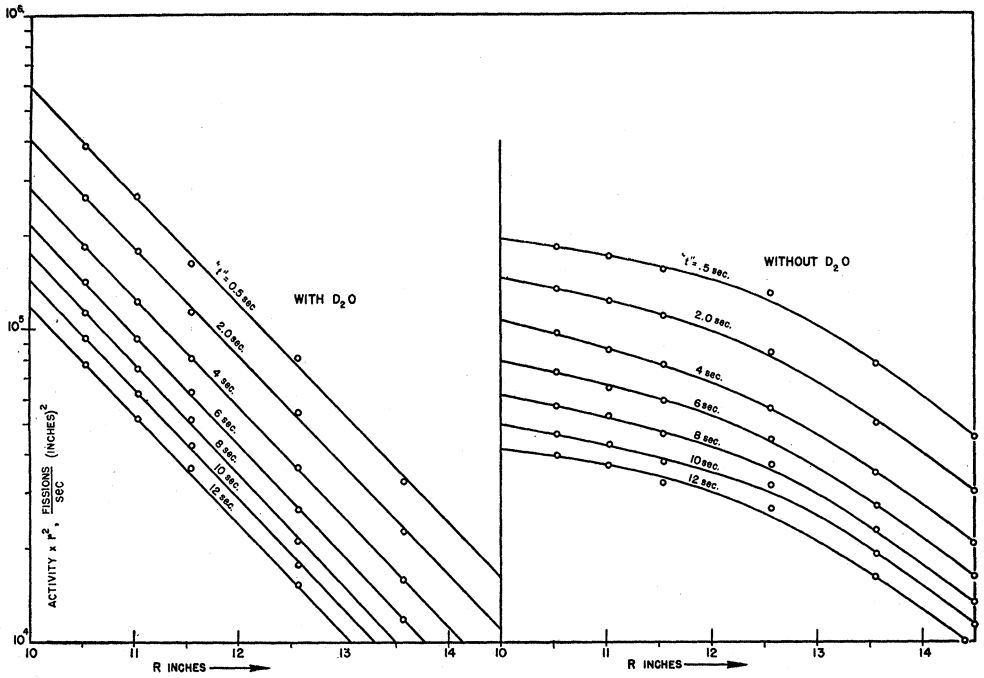

FIG. 3. Activity $\times R^2$ *vs.* R curves at different times t after the end of a 30-sec. exposure.

pile power level were found to be less than one percent over the period during which a set of data was taken, so that the use of an exposure monitor was not necessary.

For each exposure, a plot was made from the film data of the counting rate in the particular radial position as a function of t. From the group of curves forming a series, other curves were constructed of (counting rate$\times R^2$) *vs.* R, for successive fixed values of t. Here, R is the radial distance of the detector from the source at the center of the sphere. The source activity at any time is then proportional to the area under the corresponding curve.

TABLE I.* Delayed neutrons from U^{235}. τ=mean life, $\tau_{\frac{1}{2}}$=half-life, E=energy in Mev, A=saturated exposure yield relative to the yield of the 22-sec. period.

$\tau_{\frac{1}{2}}$	τ	E(Mev)	A
55.6	80.2	0.25	0.153
22.0	31.8	0.56	1.00
4.51	6.50	0.43	1.28
1.52	2.19	0.62	1.45
0.43	0.62	0.42	0.51
0.05	0.07		0.15
		Total	4.54

* Data for the first five periods from unpublished work of D. J. Hughes, J. Dabbs, and A. Cahn; for the shortest period from D. Hall.

Examples of these (counting rate$\times R^2$) *vs.* R curves are shown in Fig. 3. With heavy water in the sphere, the curves on a semi-log scale are linear with a relaxation length (inverse slope) which is constant and equal to 1.25 inches. This indicates that the neutrons have been well thermalized before they escape into the surrounding oil. When the sphere is empty, the semi-log plot of the neutron distribution shows a decided initial curvature caused by slowing down in the oil.

In general, the desired source activity due to photo-neutrons alone, at a time t_1, is the difference between the areas under the corresponding distribution curves, with and without heavy water, after correction for epicadmium activity. By computing a number of points, a curve may be drawn of source photo-neutron activity as a function of time.

We assume that the data can be represented by a formula of the form (1), where A_p is the photo-neutron activity of the source as a function of time t after the end of the exposure,

$$A_p(t)=\sum_i a_i(1-\exp-[T/\tau_i])\exp-[t/\tau_i], \quad (1)$$

T is the exposure time, τ_i the mean life, and a_i, the yield, is the activity of the ith radioactive com-

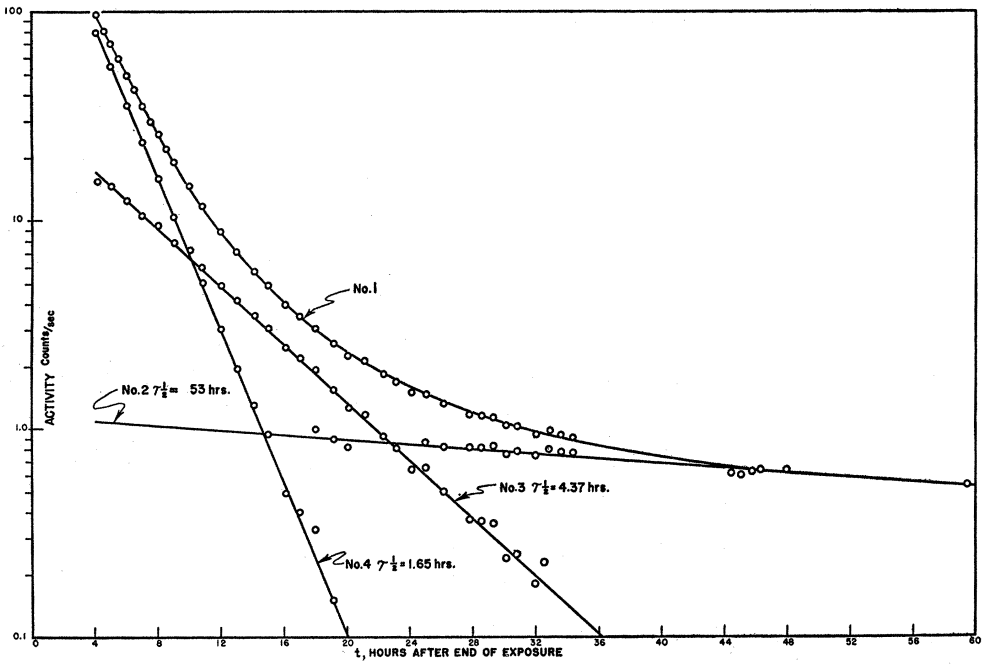

FIG. 4. 3.75-hour exposure with D_2O. Curve No. 1 shows unanalyzed data, which are decomposed into curves 2, 3 and 4 of half-lives, 53 hours, 4.4 hours, and 1.6 hours, respectively.

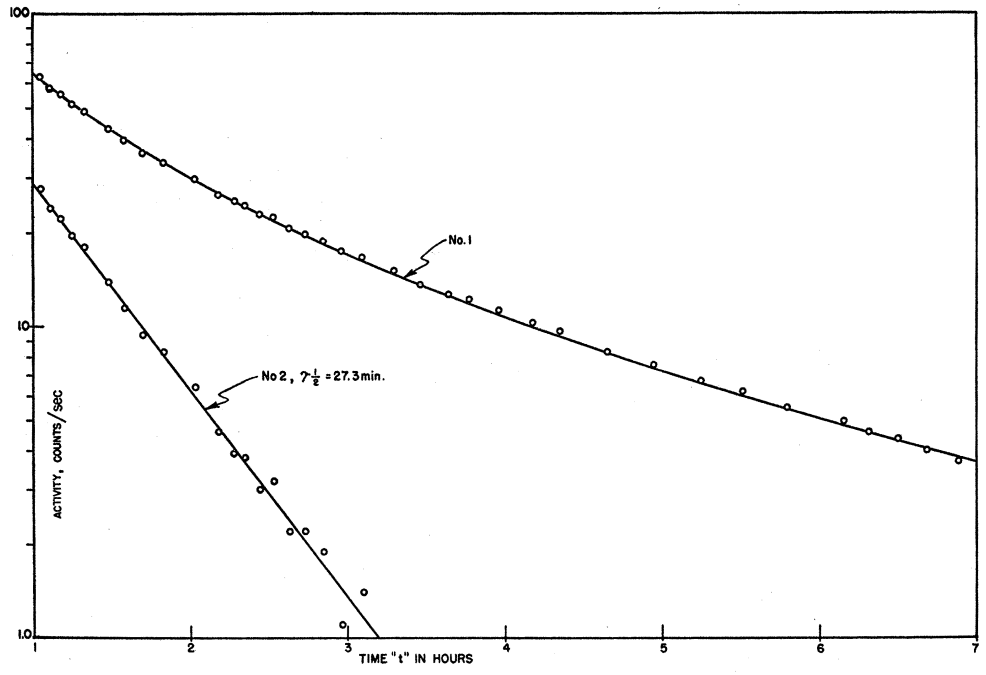

FIG. 5. Thirty-minute exposure with D_2O. Curve No. 1 shows unanalyzed data. Curve No. 2 of half-life 27 minutes is obtained by subtracting from curve 1 the activities caused by the 53-hour, 4.4-hour, and 1.6-hour periods.

ponent for $T=\infty$, $t=0$. The longest period is determined first, at large values of t when all shorter periods have died out, and its activity subtracted from the total. The shorter periods are determined, successively, by a repetition of the process. A similar formula for $A_d(t)$, the delayed neutron activity, was fitted to the decay curve taken with the sphere empty. In this case, the yields and periods[3] were taken from Table I.

[3] The yields and periods of the delayed neutrons have been studied by several groups of workers on the uranium project. One of the earliest studies is that of A. H. Snell, J. S. Levinger, R. G. Wilkinson, E. P. Meiners, and M. B.

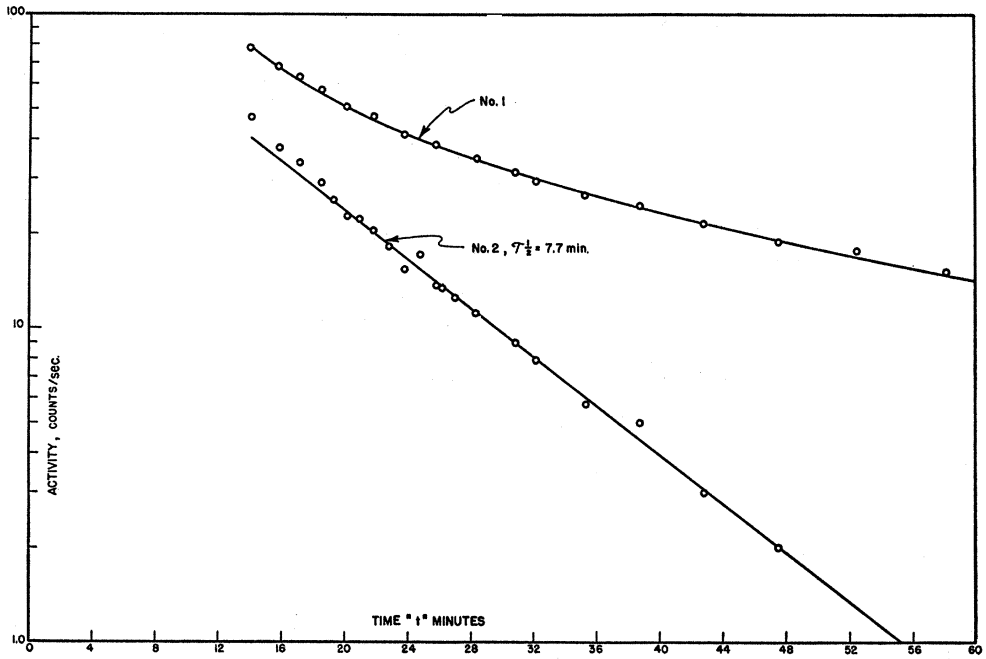

FIG. 6. Five-minute exposure. Curve No. 1 shows unanalyzed data. Curve No. 2 of half-life 7.7 minutes is obtained from curve No. 1 by subtracting the activities caused by the 4 longer periods.

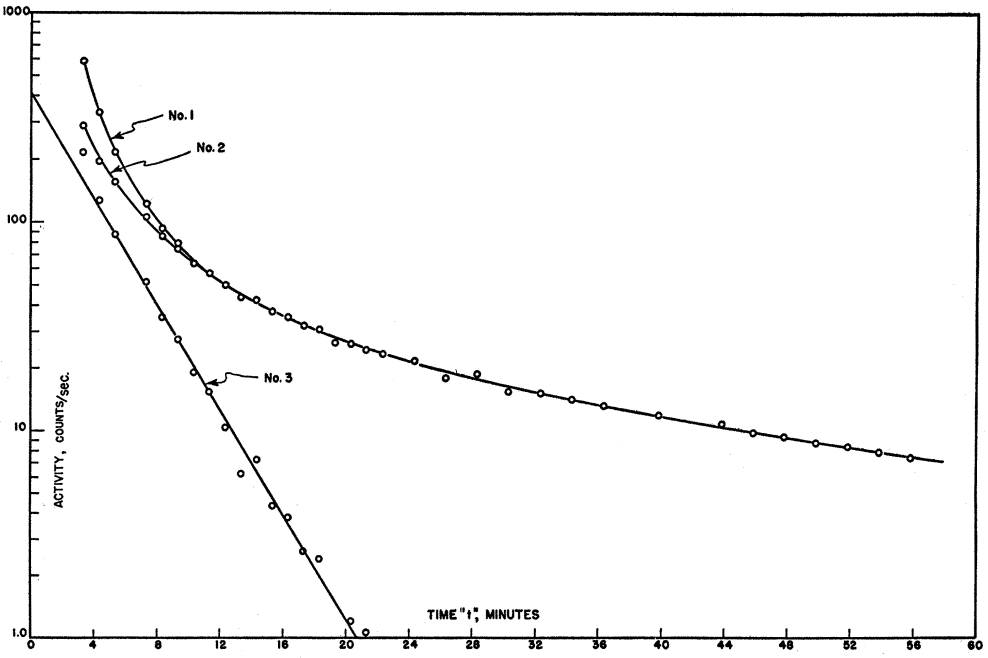

FIG. 7. Three-minute exposure. Curve No. 1 is the total activity with D_2O. Curve No. 2 is obtained from curve No. 1 by subtracting the delayed neutron activity. Curve No. 3, showing 2.4 minute half-life, is obtained from curve No. 2 by subtracting the activities caused by the 5 longer periods.

By extrapolation to $t=0$, $T=\infty$, we can then express the yields a_i of each photo-neutron period as fractions of the total delayed neutron yield $A_d(t=0, T=\infty)$.

Sampson, Phys. Rev. **70**, 111 (1946). The values in Table I were taken from more recent unpublished work of D. J. Hughes, J. Dabbs, and A. Cahn, and also from the work of D. Hall.

IV. EXPERIMENTAL RESULTS

A. Periods and Uncorrected Yields

The longest delayed neutron component has a half-life of 55 seconds, so for $t > 10$ min. the delayed neutron activity is negligible in comparison with that of the longer period photo-

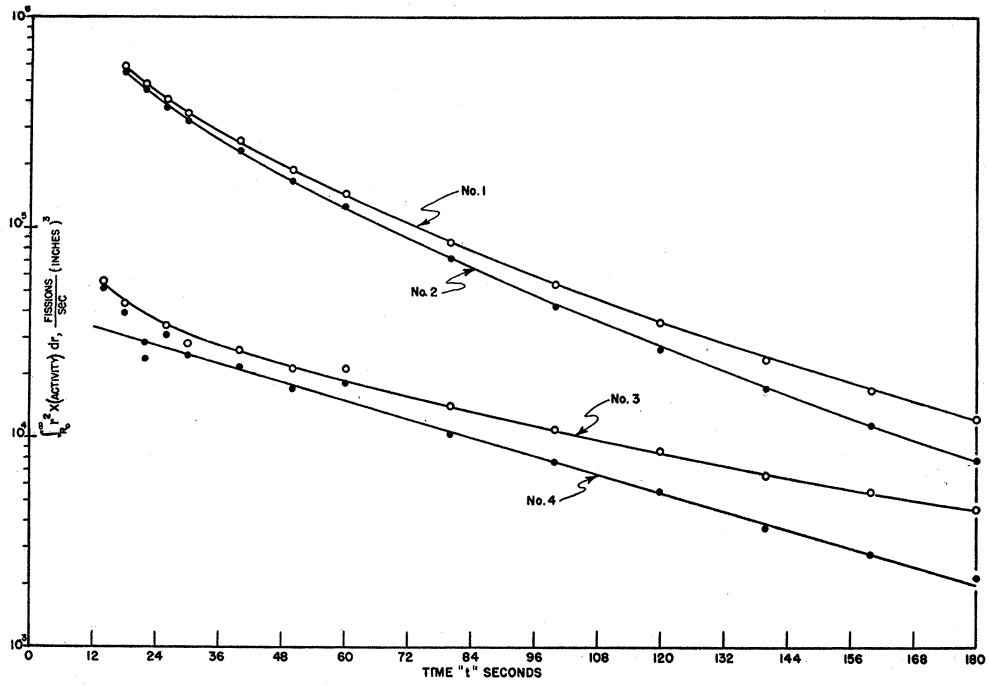

FIG. 8. Thirty-second exposure volume integrals. Curve No. 1 is the total activity with D_2O. Curve No. 2 is the total activity without D_2O. Curve No. 3, showing photo-neutrons alone, is obtained by subtracting No. 2 from No. 1. Curve No. 4, with half-life 41 seconds, is obtained from No. 3 by subtracting the activities due to the 6 longer periods.

neutrons. Under these conditions, decay curves of the gross activity with D_2O in the sphere represent photo-neutrons alone. Figures 4–6 show the gross activity from $t=15$ min. to $t=60$ hr., and the 5 component periods into which the data may be analyzed. The half-lives of these components are 53 hours, 4.4 hours, 1.6 hours, 27 minutes, and 7.7 minutes. The longest period, $\tau_{\frac{1}{2}}=53$ hr., was actually followed somewhat beyond the 60-hr. limit of Fig. 4, but its half-life is considerably less certain than those of the shorter periods.

TABLE II. Photo-neutron yields and periods.

$\tau_{\frac{1}{2}}$ = half-life, τ = mean life.
A = uncorrected photo-neutron yield relative to that of the 22-sec. delayed neutron period.
E_γ = γ-ray energy in Mev, from unpublished work of Hughes, Spatz, and Cahn.
$\frac{1}{F_\gamma}$ = factor to correct yields measured with the 10 in. radius sphere of D_2O to an infinite sphere.
α = correction for γ-ray absorption in the rabbit wall, Al tube, etc.
β = correction for multiple Compton scattering.
$A_c = A\times\alpha\beta/F_t$, yield corrected to infinite volume of D_2O.
$A_d = 100\,A_c/4.54$, percent yield of photo-neutrons relative to the total yield of delayed neutrons.
$A_f = 2\times 0.01\,A_d/100$, absolute yield of photo-neutrons per fission using the value: fast neutrons/fission = 2.
$\frac{1}{F_\gamma}$ = number of γ-ray photons per photo-neutron in an infinite volume of D_2O, assuming each collision reduces energy below threshold.
C = correction factor allowing for photons not reduced below threshold in first impact.
A_γ = absolute number of photons per fission.

Ref. No.	$\tau_{\frac{1}{2}}$	τ	A	E_γ	$\frac{1}{F_\gamma}$	α	β	A_c	% A_d	$A_f\times10^4$	$1/F_\gamma$	C	A_γ
1	53 hr.	76 hr.	0.00038	(3)	1.69	1.06	1.085	0.00074	0.0163	0.0326	476	1.14	0.0015
2	4.4 hr.	6.3 hr.	.00119	(3)	1.69	1.06	1.085	.00232	.0515	.101	476	1.14	.0042
3	1.65 hr.	2.4 hr.	.0093	2.62	1.62	1.06	1.052	.0168	.370	.742	670	1.09	.045
4	27 min.	39 min.	.0082	2.60	1.63	1.06	1.056	.0149	.328	.656	643	1.09	.038
5	7.7 min.	11 min.	.0124	3.0	1.69	1.06	1.085	.0242	.534	1.065	476	1.14	.046
6	2.4 min.	3.4 min.	.0276	2.65	1.63	1.06	1.054	.0504	1.11	2.23	650	1.09	.132
7	41 sec.	59 sec.	.090	2.25	1.52	1.06	1.01	.147	3.24	6.50	2450	1.01	1.58
8	2.5 sec.	3.6 sec.	.225	3.4	1.77	1.06	1.11	.469	10.31	20.7	389	1.18	.675
								Total→16%		3.2×10^{-3}			2.5

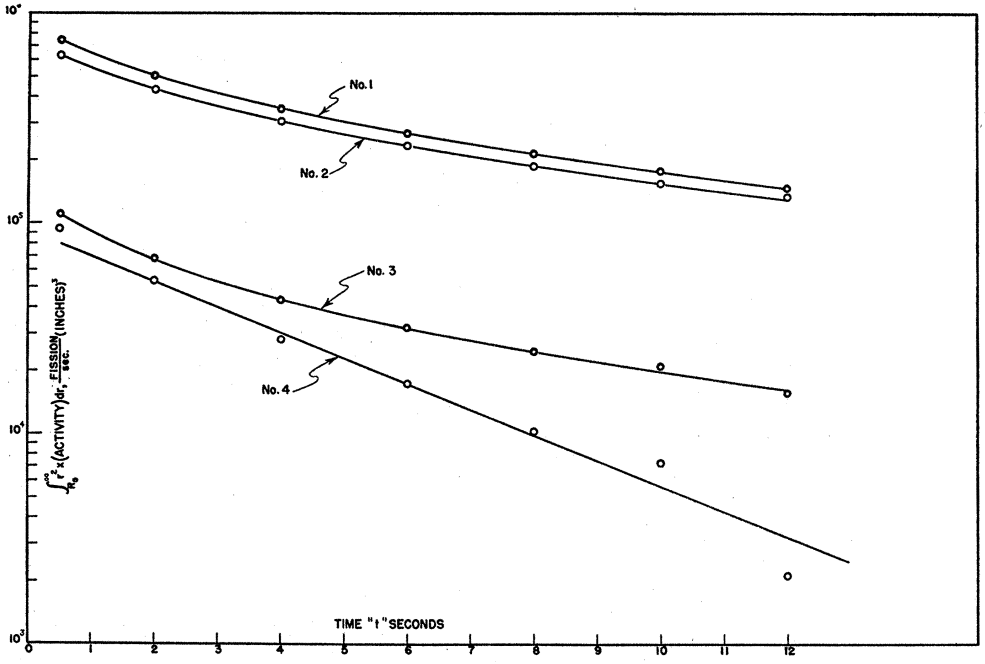

FIG. 9. Thirty-second exposure volume integrals. No. 1 is the total activity with D_2O. No. 2 is the total activity without D_2O. No. 3 is the activity caused by photoneutrons alone. No. 4, showing a half-life of 2.5 seconds is obtained from No. 3 by subtracting the activities due to the 7 longer periods.

Figure 7, curve No. 1, shows the total activity with D_2O following a 3-min. exposure. Curve No. 2 shows the photo-neutron activity alone, after subtracting from No. 1 the contribution of the delayed neutrons, determined with the sphere empty. A formula representing the 5 previously determined photo-neutron periods was fitted to curve No. 2 at $t=53$ min. and then subtracted off, giving curve No. 3. A sixth period is indicated, $\tau_{\frac{1}{2}}=2.4$ min. Extrapolating all yields back to $t=0$ and infinite exposure, the yields of the six photo-neutron periods could then be expressed in terms of the delayed neutron yields. These values are given in Table II.

In Fig. 8, curve No. 1 shows the total activity with D_2O and No. 2 the activity without D_2O, for an exposure of 30 sec. and a 0.15-g sample of enriched material. The delayed neutron curve has been corrected for epicadmium activity, which amounted to about 2 percent. The difference between curves 1 and 2 amounts to only 10 percent of the total activity at $t=30$ sec., so the experimental accuracy in determining the photo-neutron yields for the short periods is considerably less than for the longer periods. After subtracting off the longer periods, a component of half-life 41 sec. is indicated by curve No. 4.

Figure 9 gives the result of a 30-sec. exposure with a 1.5-g sample of unenriched uranium. Here the decay curve is extended back to within 0.5 sec. after the end of the exposure. A period $\tau_{\frac{1}{2}}=2.5$ sec. is found.

B. Estimated Accuracy

The accuracy to be expected in obtaining the area under one of the curves of the type illustrated in Fig. 3 is not better than one percent, owing to statistical probable errors alone. Hence, the yields given in column A, Table II, for the two shortest periods where the photo-neutron activity is of the order of 10 percent of the total are uncertain by about 15 percent. Except for the 53-hr. period, the yields of the longer periods should be considerably more accurate.

The half-life and yield of any short period depends on the previously determined constants of all longer periods, giving a cumulative error. The possibility cannot be ruled out that some of our shorter periods are fictitious, that perhaps some of them cannot be identified with individual fission isotopes. Indeed, the suggestion is made below that the shortest periods given in Table II cannot represent a single isotope but must be due to many half-lives too close together to be resolved. However, the periods and yields given

do represent the photo-neutron activity as a function of time fairly well.

If the heavy water were as pure as stated by the manufacturer, the absorption of neutrons in it would have been negligible. Since contamination might have been introduced during the course of the experiment, the neutron absorption of the heavy water was tested in the following manner: An Sb-Be photo-neutron source was placed at the center of the sphere and the neutron distribution curves in the oil were obtained, with and without heavy water. The γ-rays from Sb have energies less than 2.17 Mev and cannot create any photo-neutrons in the heavy water, so the areas under the (counting rate)$\times R^2$ *vs.* R curves should be equal in the two cases. Actually the areas were found to be the same within about 2 percent.

C. Corrections to Experimental Yields

In Table II, column A represents photo-neutron yields (saturated activities) for our sphere of heavy water relative to the 22-sec. delayed neutron period yield as 1.00. In order to correct these yields to the case of an infinitely large sphere of D_2O and complete γ-ray absorption, it is necessary to know the energy of the γ-rays. Conclusions about the initial energy of the photo-neutrons could not be made from the data of this experiment. The large amount of heavy water moderator slowed the neutrons down to such an extent that the shape of the distribution curve in the oil, where the measurements were taken, was no longer a measure of the initial neutron energy. Hence, we have tried to correlate our periods with those found by W. D. B. Spatz, D. J. Hughes, and A. Cahn in unpublished work on the same subject. Their values for the γ-ray energies are given in column E_γ, Table II. For purposes of calculation we have arbitrarily assumed an energy of 3 Mev for the two longest periods, which were not reported by Spatz, Hughes, and Cahn.

In D_2O and for γ-ray energies between 2 and 5 Mev the photoelectric effect and pair production are negligible in comparison with the Compton effect. If we assume that every Compton scattering of a γ-ray quantum by an electron reduces the quantum energy below the γ,n threshold in D_2O, the fraction of the quanta absorbed in a sphere of radius R cm, from a central point source, is $F_t=1-\exp-[(\mu_s+\mu_\gamma)R]$ where μ_s is the linear "absorption" coefficient per cm for Compton scattering and μ_γ is the coefficient for the γ,n reaction in D_2O. For this experiment the effective thickness of heavy water is 23.3 cm. $\mu_s=\sigma_s\times 3.35\times 10^{23}$ per cm^3 for D_2O, where σ_s, the Compton scattering cross section per electron varies from about 1.27×10^{-25} cm^2 at 2.5 Mev to 1.06×10^{-25} at 3.5 Mev. $\mu_\gamma=6.7\times 10^{22}\times\sigma_\gamma$ per cm^3, where σ_γ, the γ,n cross section per deuteron varies from zero at 2.17 Mev to about 14×10^{-28} cm^2 at 3.5 Mev. In Table II are listed values of $1/F_t$ the correction factor by which the yields A must be multiplied to give the yields for an infinite sphere of D_2O, for the assumed energies E_γ.

A small correction, $\alpha=1.06$, is listed in the next column of Table II. This compensates for the γ-ray absorption in the 0.050-inch wall thickness of the pneumatic tube, the fixed 0.125-inch thick Al tube through the center of the sphere, and in the $\frac{3}{16}$-inch thick wall of the plastic rabbit.

Column β in Table II gives an additional correction necessary because not every Compton scattering process reduces the energy of a γ-ray quantum below the γ,n threshold, so that the flux of γ-rays of energy above the threshold is everywhere higher than that calculated using the full Compton scattering cross section. β/F_t is then the ratio of the number of γ,n neutrons produced by multiple scattered photons in an infinite sphere of D_2O to that produced by multiple scattered photons in a sphere of radius 23.3 cm. The theory of the multiple scattering correction was developed for us by Mr. E. Greuling and Mr. H. Soodak.

Column A_c in Table II is $A\times\alpha\beta/F_t$, the corrected yield of photo-neutrons in an infinite D_2O medium, relative to the yield of the 22-sec. delayed neutron period. $A_d=100\times A_c/4.54$ is the percent yield relative to the total delayed neutrons (see Table I, column A). Column $A_f=2\times 0.01\times A_d/100$ is the absolute yield of photo-neutrons per fission, calculated from the data in column B, Table I, using the arbitrary value 2 for the number of instantaneous neutrons

emitted per fission of a U^{235} nucleus and 0.01 for the ratio[4] of delayed to instantaneous neutrons.

D. Gamma-Ray Yields

We now compute the number of γ-rays per fission necessary to produce the photo-neutrons of each period, with yields A_f in Table II. The fraction of the total γ-ray quanta from a point source which are absorbed in the γ,n reaction in an infinite D_2O sphere is

$$F_\gamma=\mu_\gamma/(\mu_\gamma+\mu_s),$$

provided each scattering process is assumed to reduce the energy below the threshold. If we take into account the fact that some scattered quanta still have energy above the threshold, the actual photo-neutron production will be higher by a factor C, which will be small near the threshold and increase with the initial energy of the γ-ray. In Table II are listed, for each period, the values of $1/F_\gamma$ and of C. The absolute yield of γ-ray quanta per fission, A_γ, will be

$$A_\gamma=A_f/F_\gamma C.$$

V. DISCUSSION OF RESULTS

A. The Effect of Possible Parent-Daughter Contributions

In the analysis of the data as described above it has been assumed that the hard γ-ray emitters are initial fission fragments. The interpretation of the data is made uncertain because of the possibility that the emitter of the photon giving the photo-neutron may be a daughter of the initial fragment. In such cases the data would yield the true period, but the amplitude deduced by extrapolating the decay back to zero time may be in error. If the daughter has a very much longer life than its parent, the error will not be large. If the two have comparable lifetimes the saturated activities given may be a considerable overestimate.

B. Number of Photons per Fission

From the number of photo-neutrons per fission, the number of hard photons per fission was calculated from a consideration of the relative probabilities of Compton and γ,n collisions. The results are given in column A_γ of Table II. The sum of the values in this column is 2.5 photons per fission of energy above 2.17 Mev. The highest single yield is 1.6 for the 41-sec. period. This value, however, is subject to great uncertainty. The assumed energy $E_\gamma=2.25$ Mev is very near the threshold, where the γ,n cross section is small and changing rapidly. If the true energy is only slightly higher, the calculated γ-ray yield A_γ would be much reduced. However, the highest observed yields of individual fission products are only about 6 percent, so the γ-ray yields of the 2.5-sec. 41-sec., and 2.4-min. periods are all much too high to be caused by single isotopes. These high photon yields can be explained by supposing that there are a large number of high energy γ-emitters with half-lives too close together to be resolved in this experiment.

C. Correlation of the Gamma-Ray Periods with Known Fission Products

We have been unable to correlate without ambiguity our periods with those given for the known fission products. It seems that in order to accomplish this correlation it would be necessary to repeat the experiment with individual fission products or with groups of products each containing a small number of members.

[4] H. D. Smyth, *Atomic Energy for Military Purposes* (Princeton University Press, Princeton, 1945), Appendix 3; or Rev. Mod. Phys. **17**, 459 (1945).

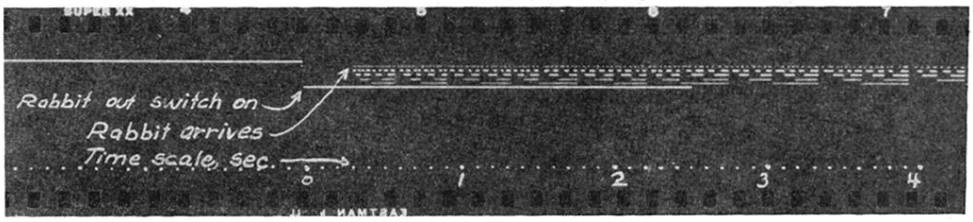

FIG. 2. Sample of film recorded data.