Delayed Neutrons Which Accompany Photofission of Uranium and Thorium

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Decay curves and yield were obtained from delayed neutron radiation accompanying the photofission of uranium and thorium. The delayed neutrons represent 0.41 ± 0.02% and 0.18 ± 0.01%, respectively, of all the neutrons emitted in photofission of uranium and thorium.

AT the present time, there are many papers available devoted to the investigation of delayed neutrons which accompany the fission of U^{236}, U^{239} and Th^{233} isotopes, produced during irradiation of uranium and thorium with neutrons. There is in the literature no study of delayed neutrons in photofission of uranium and thorium, where fission of the primary isotopes of U^{235}, U^{238} and Th^{232} takes place. In 1950 such a work was performed in the laboratory of V. I. Veksler on samples of natural uranium and thorium.

Photofission cross sections of U^{235} and U^{238} differ from each other by a factor of 1.5-2[1,2]. Therefore, the delayed neutron radiation obtained by irradiation, with x-rays, of uranium sample composed of the natural isotopic mixture pertains almost entirely to the U^{238} isotope, the content of which is 99.3% in natural uranium.

Measurements were performed with the synchrotron at the x-ray energy $E_{max} = 18.5$ mev. The mean excitation energy of fission nuclei,

$$E = \int_{E_{thr}}^{E_{max}} E\sigma_f(E) f(E)\, dE \Big/ \int_{E_{thr}}^{E_{max}} \sigma_f(E) f(E)\, dE$$

[E is the energy of photons; $f(E)$ is the number of photons having energy $E + dE$; σ_f is the photofission cross section; E_{thr} is the energy corresponding to the photofission threshold], calculated in accordance with available curves obtained by I. V. Chuvilo, which represent photofission cross sections as a function of photon energy, is equal to 12-12.5 mev, for uranium and thorium, at $E_{max} = 18.5$ mev.

The irradiated samples of uranium and thorium, in the form of discs 4 cm in diameter, were placed in the center of a large paraffin block. The γ-rays striking the samples passed through a round canal 5 cm in diameter made in paraffin. The delayed and the "instantaneous" neutrons ejected at the instant of photosplitting, were registered in the ionization chamber KH-14, filled with BF_3 after having been slowed down in paraffin. In Fig. 1 are illustrated the plots of the decreasing activity of delayed neutrons, which accompany the photofission of uranium and thorium, against time. The curves were obtained from a great number of sample irradiation cycles followed by the registration of neutrons. In each cycle, samples were irradiated with γ-rays for 3 min, then the irradiation was interrupted and after 1/2 sec, the registration of neutrons started and was continued for 5 min. From Fig. 1 it is seen that in the case of uranium, and thorium as well, the activity of delayed neutrons decreases rapidly with time. The character of this decrease of neutron activity indicates that most of the delayed neutrons have decay half-periods of less than 30 sec.

The exact determination of the decay periods and their relative yields was difficult due to the small cross section of the photofission. For uranium the following decay half-periods were established:

$$T_1 = 58 \text{ sec} \pm 8 \text{ sec}; \quad T_2 = 22 \text{ sec} \pm 3 \text{ sec};$$
$$T_3 = 5.5 \text{ sec}; \quad T_4 \sim 2 \text{ sec}.$$

The yield of delayed neutrons with a 22 sec period was several (about six) times greater than the yield of neutrons with the period of 58 sec. The correlation of the periods obtained, with the half-decay periods of the delayed neutrons which accompany the fission under the action of

[1] J. McElhinney and W. E. Ogle, Phys. Rev. 81, 342 (1951)

[2] J. R. Huizenga, J. E. Gindler and R. B. Duffield, Phys. Rev. 95, 1009 (1954)

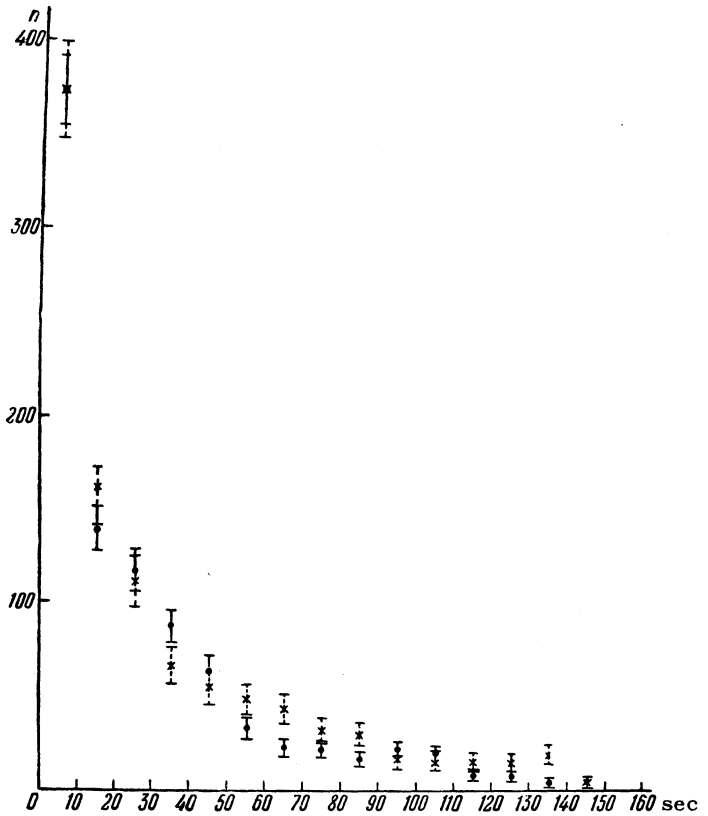

FIG. 1. Decay curve for delayed neutrons of photofission of uranium and thorium. ● – uranium, × – thorium, n – number of delayed neutrons.

neutrons [3,4] permits us to assume that in photofission there are formed the same neutron-active nuclei as in the case of neutron-induced fission.

YIELD OF DELAYED NEUTRONS

a) *Method of measurement.* The yield of delayed neutrons relative to the total yield of photoneutrons emitted in photofission of uranium and thorium was measured by a method which could be applied because of the pulse character of the accelerator. The synchrotron with which the measurements was made, produces γ-pulses of about 20 μ-sec duration, and frequency of 150 pulses/sec (the interval between γ-pulses being 6,667 μ sec). A special electronic circuit was developed which allowed the neutron pulses leaving the ionization chamber to be recorded in various time intervals between γ-pulses. Pulses from the ionization chamber were fed into a preamplifier consisting of two amplifying stages. Through the inverting stage with a double triode, 6N7, pulses were transmitted from the preamplifier in opposite phase, using a double-shielded symmetric twin-conductor cable (in order to reduce the amount of pick-up of electric disturbances), to the main amplifier, where they were recombined in proper phase.

All three preamplifier tubes were mounted in the lower part of a pipe, 5 cm in diameter, at the end of which, chamber KN-14 was placed. This setup was very convenient in working with a paraffin block. As the main amplifier, type ZH was used (amplification band up to $3 - 5 \times 10^6$ cps) with its agram slightly changed to suit the given problem. In order to decrease the charging time of the amplifier, at the instant of γ-pulse, a relatively small resistance of the order of 1 MΩ was included in the chamber circuit. For the purpose of decreasing recovery time of the system, which is dependent on grid currents during charging of the amplifier, the

[3] D. J. Hughes, J. Dabbs, A. Cahn and D. Hall, Phys. Rev. 73, 111 (1948)

[4] K. H. Sun, R. A. Charpie, F. A. Pecjak, B. Jennings, J. F. Nechaja and A. J. Allen, Phys. Rev. 79, 3 (1950)

time constant, *RC*, of grid circuits of the amplifier tubes was also reduced to 1 μsec. Moreover, in order to eliminate the secondary pulses, which were obtained due to multi-differentiation, diodes were included in two stages, which cut off the pulses from repeatedly entering.

Thus, the system recovered within a period of time of the order of 1 μsec. After amplification, pulses from the ionization chamber passed through a control circuit, resulting in time selection of pulses. The working principle of the control circuit was the following:

1) Pulses from the amplifier entered the grid of the input tube and then were transferred through a noise reducing diode into a fast counting system.

2) The diode was closed or opened depending on the voltage on the control grid of the special modulating tube which was also connected to the anode of the input tube.

3) Associated with the instant of the exclusion of high-frequency voltage on the "endo"-vibrator and preceeding each γ-pulse, the control pulse which is connected with the moment of switching off the high-frequency voltage on the vibrator, and which precedes each γ-pulse, traversing the system of vibrators, there is applied to the grid of the modulating tube a selective rectangular pulse, the width and position of which, relative to the control pulse, could be changed by means of potentiometers in the circuits of univibrators.

4) The diode was open for pulses from the amplifier only at the instant of arrival of the selective pulse. Pulses from the output of the diode were fed to the fast counting system throughout the duration of the rectangular pulse. The duration of the rectangular pulse--a measurable interval--determined the "gating" of the arrangement, and its position relative to the γ-pulse (the initial rise) determined the "delay" relative to the γ-pulse. The gating and delay were established by means of the timing pips on the ocsilloscope and could be changed within rather wide limits, from 50 to 7000 μ sec.

5) The by-pass capacitor of the main diode, through which the differentiated fronts of the selective pulse had to pass, was compensated by means of a second selective pulse which had opposite phase. This pulse was passed through the by-pass capacitor of the auxiliary diode (second half of the envelope) into the output circuit of the main diode.

The time distribution of the number of instantaneous photoneutrons in lead upon photodisintegration is plotted in Fig. 2. The abscissa gives the time in microseconds, and the ordinate gives the logarithm of the counter number of neutrons, measured at constant gating of 200 μ sec, and with various delays. As follows from Fig. 2, the neutron density decreases with time according to the law e^{-t/τ_0}, where $\tau_0 = 183 \pm 3$ μ sec. After a time $t = 2000$ μ sec, the photoneutrons are practically completely absorbed in the paraffin. Therefore, the "instantaneous" neutrons were recorded during the first 2000 μ sec after the γ-pulse. To eliminate the strong pulse of γ-rays, the instantaneous neutrons were recorded with a 20 μ sec delay. Correction was made for the number of neutrons in the first 20 μ sec after the γ-pulse.

Table I gives the number of neutrons recorded at the various delays indicated in the first line, with a constant gating of 500 μ sec for uranium, thorium and lead. The number of the instantaneous neutrons of lead, recorded with this arrangement, drops by a factor of 15 when the delay is changed to 500 μ sec, which is in complete agreement with the measured lifetime of neutrons in paraffin. As a consequence of the rapid decrease in the intensity of the neutrons, about 94% of the total number of neutrons are recorded in a 500 μ sec delay. Only 0.03% are recorded for 1540 μ sec delay, while for a 2000 μ sec delay, the counted number of neutrons drops to zero.

A similar drop of the instantaneous neutrons with time was obtained by us for copper and zinc.

The initial intensities for all three samples--uranium, thorium and lead--were of the same order; therefore, because of the absence of delayed neutron radiation in uranium and thorium, neutrons should not have been recorded after 2000 μ sec from the γ-pulse, just as in lead. However, it is seen from Table I that, from the 2000 μ sec time to the next γ-pulse, constant neutron activity was observed for uranium and thorium, corresponding to delayed neutron radiation. Simultaneous recording of the neutrons in the first 2000 μ sec after the γ-pulse and of the delayed neutrons in the interval from 2000 to 6000 μ sec after the γ-pulse allows the determination of the ratio of the delayed neutrons to the total number of photoneutrons. This method of recording the neutrons is very effective, since it permits the recording of the delayed neutron radiation for 2/3 of the time at saturated activity, which yields a several fold increase in the statistics. Large samples had to be used because of the high background of instantaneous neutrons from the synchrotron and

TABLE I

Delay (in μsec)	40	540	1040	1540	2040	2540	3040	3540	4040	4540	5040	5540	6040
Uranium	65520 ∓230	4622 ∓50	309 ∓17	56 ∓6	43 ∓2	42 ∓3.5	41 ∓3.5	42 ∓3.5	48 ∓3.7	40 ∓3.5	45 ∓5	40 ∓5	47 ∓2.5
Thorium	20952 ∓105	1394 ∓28	89 ∓7	8 ∓1.7	5 ∓0.9	6 ∓1.2	8 ∓1.4	5 ∓1.1	5 ∓1.1	6 ∓1.2	6 ∓1.3	6 ∓1.2	6 ∓1.2
Lead	28023 ∓109	1729 ∓33	120 ∓8	8 ∓2	1 ∓1	0	0	0	0	0	0	0	0

because of the small output of delayed neutrons. In order to maintain the same conditions of neutron absorption in background determination, the samples were placed in small cadmium boxes (wall thickness ½ mm), the same empty cadmium box being put in place of the sample in background measurements. The sample of uranium weighed 284 gm, that of thorium 120 gm. The background of instantaneous neutrons amounted to ~ 10% in the uranium case and ~ 20% for thorium. In the measurement of delayed neutrons, the background was practically equal to zero, since the neutron background from spontaneous fission was negligibly small.

Comparison of the number of incident neutrons recorded in a 500 μ sec interval and for various delays from 2000 μ sec to 6000 μ sec (Table I) shows that the intensity of the radiation of the delayed neutrons is constant to within a few percent in the time interval from 2000 to 6000 μ sec. This means that there is no appreciable number of nuclei with a decay period of the order of a few milliseconds among the neutron active nuclei formed during photo-fission.

Yields of instantaneous and delayed neutrons were measured for a fixed position of the ionizatior chamber in paraffin. In the calculation of the ratio of the delayed neutrons to the instantaneous neutrons, corrections were made for the different space distributions of the densities $\rho(r)$ of the delayed and instantaneous neutrons of uranium. The curves in Fig. 3 show the spatial distribution $\rho(r)\,r^2$ for delayed and instantaneous neutrons of uranium. Similar curves were obtained for thorium. The curves meet at 5 = 10 cm, where the chamber was placed for the measurements. As is seen from these curves, the spectrum of the delayed neutrons is appreciably softer. The curves that have been obtained permit us to estimate the mean energy of the delayed and instantaneous neutrons.

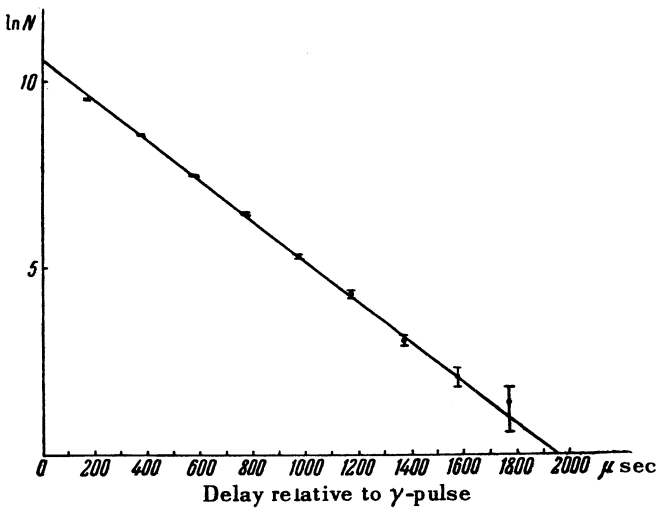

FIG. 2. Number of photoneutrons of lead N, recorded in paraffin, at constant gating (200 μsec) and various delays relative to the γ-pulse; $\tau_0 = 183 \pm \mu$sec.

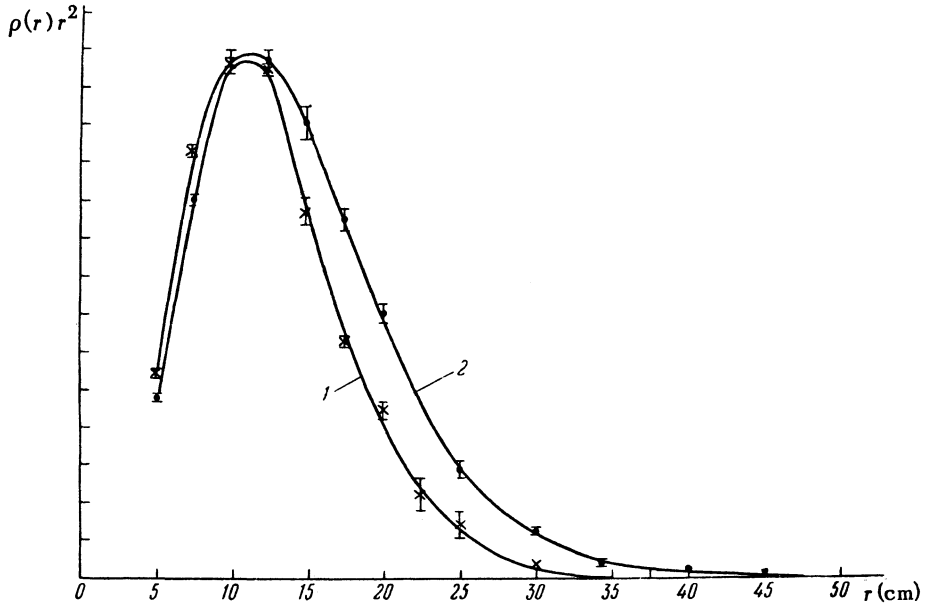

FIG. 3. Space distribution of the delayed and instantaneous neutrons of uranium in paraffin. *1.* delayed neutrons, *2.* instantaneous neutrons.

TABLE II

	Source (Ra_α+Be)	Photoneutrons of uranium	Photoneutrons of thorium	Delayed neutrons of Uranium	Photoneutrons (Na_γ^{24}+Be)
$\overline{r^2}$, cm^2	362	259	243	185	199
$\sqrt{\overline{r^2}}$, cm	19.0	16.1	15.6	13.6	14.1
Mean energy, mev	5 4	2.5 2.1	2.1 1.8	0.4 0.5	0.83∓0.04

Table II lists the mean square distance from the sample, which was the neutron source, for instantaneous neutrons of uranium and thorium, and delayed neutrons of uranium. In the same Table the values of the quantity $\overline{r^2}$ are given for photoneutrons of a (Na_γ^{24} + Be) source and (Ra_α + Be) source, measured for calibration purposes. The (Na_γ^{24} + Be) source emits monochromatic neutrons with energies 0.83 ± 0.04 mev.

The mean neutron energy of the source (Ra_α + Be) is 4-5 mev according to various data in the literature. The mean energy of the instantaneous neutrons of uranium and thorium is about 2 mev and of the delayed neutrons of uranium, ~ 0.5 mev. The measured mean energy of the delayed neutrons, brought about by photofission of uranium, coincides with the mean energy of the delayed neutrons emitted in the fission of U^{235} under the action of slow neutrons (515 ± 60 kev).

a) *Yield of delayed neutrons.* For uranium, the yield of delayed neutrons, relative to all neutrons emitted in photodisintegration, amounted to 0.41 ± 0.02%; for thorium, 0.18 ± 0.01%.

In the photodisintegration of uranium and thorium by γ-bremsstrahlung with E_{max} = 18.5 mev, four reactions involving neutron emission are energetically possible: (γ, f), (γ, n), (γ, nf) and (γ, $2n$). Thus the yield of delayed neutrons is larger relative only to the neutrons which accompany photofission. In 1951-1952, measurements were made, with this same arrangement, on the average number of neutrons ν, relative to a single fission, in the

photodisintegration of uranium and thorium by γ-rays with $E_{max} = 18.5$ mev[5]. The following values of ν were obtained:

uranium 6.2 ± 0.5;

thorium 14.2 ± 1.2.

If in accord with the results of this research, we assume the average number of neutrons emitted immediately in photofission to be equal to 3, then the percent of delayed neutrons (relative to instantaneous neutrons of photofission for U^{238} and Th^{232} at a mean excitation energy of ~ 12 mev) is equal to $\sim 0.8\%$.

[5] B. N. Valuev, B. I. Gavrilov, G. N. Zatsepina and L. E. Lazareva, J. Exper. Theoret. Phys. USSR **29**, 280 (1955); Soviet Phys.

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