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**MEASUREMENT OF DELAYED NEUTRON
YIELDS IN PLUTONIUM, URANIUM-233,
URANIUM-238, AND THORIUM RELATIVE
TO YIELD IN URANIUM-235**

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August 1955

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IDAHO DIVISION

August, 1955

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ABSTRACT

Delayed neutron studies have been made in the Experimental Breeder Reactor (EBR), using a conventional sample transfer system and a neutron counter comprised of BF_3 tubes in a graphite geometry. Samples of U^{235} , U^{233} , U^{238} , Pu and Th were irradiated in a fast flux; samples of U^{235} , U^{233} , and Pu were irradiated in a thermal flux. The following ratios of delayed neutron yields per fission (based on the longest four periods) were measured:

$$\frac{\text{U}^{235} \text{ thermal}}{\text{U}^{235} \text{ fast}} = 1.017 \pm 11\%$$

$$\frac{\text{U}^{238} \text{ fast}}{\text{U}^{235} \text{ fast}} = 2.23 \pm 7.5\%$$

$$\frac{\text{Pu fast}}{\text{U}^{235} \text{ fast}} = 0.405 \pm 7.5\%$$

$$\frac{\text{Th fast}}{\text{U}^{235} \text{ fast}} = 3.09 \pm 17\%$$

$$\frac{\text{Pu thermal}}{\text{U}^{235} \text{ fast}} = 0.360 \pm 7.5\%$$

$$\frac{\text{U}^{233} \text{ thermal}}{\text{U}^{233} \text{ fast}} = 0.956 \pm 6\%$$

$$\frac{\text{U}^{233} \text{ fast}}{\text{U}^{235} \text{ fast}} = 0.414 \pm 7.5\%$$

$$\frac{\text{Pu thermal}}{\text{Pu fast}} = 0.888 \pm 6\%$$

$$\frac{\text{U}^{233} \text{ thermal}}{\text{U}^{235} \text{ fast}} = 0.396 \pm 7.5\%$$

The divergence from unity in the case of $\frac{\text{Pu thermal}}{\text{Pu fast}}$ appears to result in part from the 5% Pu^{240} fraction in the sample used.

GENERAL

During the period September 1954 to June 1955, measurements were made in the Experimental Breeder Reactor (EBR) of the yield of delayed neutrons from the thermal fission of U^{233} and Pu, and the fast fission of U^{233} , Pu,

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U^{238} and Th relative to U^{235} . A conventional rabbit sample transfer system was used to transport samples between the reactor and the counter. The output of the counter was displayed, with a time base, on an oscilloscope and photographed.

Irradiation was accomplished in two positions in the reactor: one as near the center as possible and one in a thermal region.¹ The fuel rod size thimble being too small to admit a rabbit tube and leave clear an annular space for air, the best position that could be used was one of the inner blanket rod positions (Hole 238 was used; choice was limited to those few rod positions accessible through the top reactor shields). It was desirable that the rabbit system could be easily connected to either position. The position used for thermal irradiation was a vertical hole extending down into the graphite reactor reflector; the axis of this hole is approximately $31\frac{1}{2}$ inches from the center of the reactor and lies in the same direction from the center as Hole 238. In each case irradiation was made at the horizontal mid-plane of the reactor core. The flux in the "thermal" thimble is largely thermal, being characterized by a cadmium ratio of 11 with respect to fission of U^{235} . The flux in the "fast" thimble has a spectrum whose average energy is somewhat less than that of the spectrum shown in Figure 1, which is the measured spectrum for the center of the reactor.²

A preliminary neutron survey of the reactor area led to the selection for low background of a balcony situated about 50 feet horizontally from the thimble position as the location of the delayed neutron counting geometry; this resulted in a rabbit transit distance of approximately 80 feet. Later, it was found feasible to place the counting geometry to one side at the top of the reactor, reducing the rabbit's travel to about 30 feet. (The design of the reactor is such that the rabbit had to travel nearly 16 feet before emerging from the top shield.) Consequently, transit times for the rabbit were long and prevented any reasonable attempt to analyze delayed neutron groups having half lives shorter than about 1.5 seconds.

¹For a description of EBR, see W. H. Zinn, H. V. Lichtenberger and M. Novick, "Feasibility Report: Fast Neutron Pile for a Test of Conversion," ANL-4356 (October 14, 1949)

²H. V. Lichtenberger, et.al., "Operating Experience and Experimental Results Obtained from a NaK-Cooled Fast Reactor," Paper No. 813, International Conference on the Peaceful Uses of Atomic Energy (Geneva, Switzerland; August, 1955)

Figure 1

Neutron Spectrum from Hole #1 (Center) of the Core of the EBR.

MECHANICAL EQUIPMENT

The rabbit design used is shown in Figure 2, together with the sample in the cylindrical can. The maximum diameter of the rabbit is about 0.530 inch. The body was of aluminum and the caps of steel. The samples (Table I) were canned in stainless steel Type 347 to outer dimensions of 0.312 x 1.1 in. by Leroy Kelman of ANL Metallurgy Division. When the rabbit was assembled the sample was held under compression by the two springs shown; this was done to reduce upsetting of the sample, which occurred in early trials.

The rabbit tube was standard 5/8 in. O.D. steel tubing with 0.035 in. walls, giving an inner diameter of 0.555 in. The loose fit of the rabbit plus the reduction in diameter along the center section permitted operation through bends in the rabbit tube of reasonable radius (~ 5 ft), even though some flattening occurred in bending the tube.

A sketch of the sample transfer system is shown in Figure 3. The two thimbles used were standard EBR large thimbles (0.847 in. I.D. - 0.918 in. O.D. tube with an end fitting to position it in a blanket rod hole). These were spliced to bring the end above the reactor shield. Top fittings on these thimbles admitted the rabbit tube through an O-ring packing and at the same time provided an air hose connection to the annular space. Air was supplied from the ballast tank through a four-way solenoid valve, which could apply pressure to either end of the rabbit tube while at the same time venting the other end to the atmosphere.

The major problem was that of achieving short transit times without damage to the rabbit on arrest. Several types of dashpots were tried, but none were found capable of reliable operation over the many cycles necessary for these experiments. Ultimately a heavy, square section spring was used to absorb the impact, even though this introduced a bothersome bounce at each end of the trip. Air pressure was $\sim 7\frac{1}{2}$ psi and transit times were ~ 2.3 seconds for the first position used and ~ 1.3 seconds for the second position at the top of the reactor. Figure 4 shows the equipment on the top of the reactor.

When the counter was being operated at the more remote position (on the balcony) an AC bridge was used in conjunction with a winding around the rabbit tube at the position of irradiation to indicate the presence of the sample, thus avoiding the possibility of unknowingly having a rabbit stuck at some bend in the long tube. This was found unnecessary when operating from the top of the reactor; the impact of the rabbit at each end of its travel was clearly audible.

"Rabbit" Shown Complete and Disassembled.

Figure 2

Table I

DELAYED NEUTRON SAMPLES

Sample #	Net Weight (grams)	Analysis (%)				Form of Active Material
1	1.4439	Pu ²³⁹	94.92	±	0.05	Metal
		Pu ²⁴⁰	4.75	±	0.05	
		Pu ²⁴¹	0.315	±	0.003	
		Pu ²⁴²	0.010	±	0.001	
2	1.5050	U ²³⁵	93.38	±	0.05	Metal
		U ²³⁸	5.58	±	0.05	
		U ²³⁴	1.04	±	0.01	
3	5.2795	Th ²³²	100.00			Metal
		Th ²³⁰	<0.003			
4	10.1516	U ²³⁸	99.9666	±	0.0006	Metal
		U ²³⁵	0.0334	±	0.0006	
5	0.7766	U ²³³	98.33	±	0.02	U ₃ O ₈ Powder
		U ²³⁴	0.127	±	0.002	
		U ²³⁵	0.0102	±	0.0004	
		U ²³⁸	1.53	±	0.02	
6	0.0438	U ²³⁵	93.38	±	0.05	U ₃ O ₈ dispersed in powdered Al Vol. = .713 cm ³
		U ²³⁴	1.04	±	0.01	
		U ²³⁸	5.58	±	0.05	
7	0.0438	U ²³³	98.33	±	0.02	U ₃ O ₈ dispersed in powdered Al Vol. = .713 cm ³
		U ²³⁴	0.127	±	0.002	
		U ²³⁵	0.0102	±	0.0004	
		U ²³⁸	1.53	±	0.02	
8	0.0450	Pu ²³⁹	94.92	±	0.05	PuO ₂ dispersed in powdered Al Vol. = .713 cm ³
		Pu ²⁴⁰	4.75	±	0.05	
		Pu ²⁴¹	0.315	±	0.003	
		Pu ²⁴²	0.010	±	0.001	

Samples 6, 7, and 8 were used in the thermal flux to minimize the effect of self-shielding.

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Figure 3

Sample Transfer System.

Figure 4

Equipment in position on top of reactor. Air hose in foreground has yet to be connected to other thimble for operation. The large pipe stand around each thimble is for the purpose of absorbing the shock on the rabbit tube through the collar which is fastened around the tube just above the stand.

NUCLEAR AND ELECTRONIC EQUIPMENT

The delayed neutron counting geometry is shown in Figure 5. It consisted of a volume of graphite, 28 in. x 28 in. x 20 in., having a one-foot cube of lead at the center to shield the BF_3 counters from fission product gammas. The rabbit in counting position was at the center of the geometry. The whole was covered with 0.030 in. cadmium sheet and contained in a welded steel box for handling. Counting was done with eight BF_3 tubes, each with its own preamplifier, as shown; they were connected four in parallel in each of two recording channels.

Figure 6 is a schematic of the electronic equipment. Figure 7 is a photograph of the same equipment. No unusual equipment was used, except the mixer which was built for this specific purpose. In brief, each scaler saw the amplifier output of the four counters in its channel. In turn a signal was picked off the scaler optionally at each 10, 100, or 1000 events counted. An audio oscillator supplied 100 cycle sine wave to the mixing chassis. This sine wave was counted by a two-decade scaler, either decade of which could furnish a signal at the option of the operator. The mixer received the signals from the two neutron counting scalers, inverted one of them, and applied them to the X-terminals of the oscilloscope together with the 100 cycle sine wave and a timing pulse from the scaler strips in the mixer chassis every 10 or 100 cycles. No Y-motion was used; the signal was photographed by a Fairchild oscilloscope camera with shutter always open and film moving continuously in the vertical direction. Film speeds ranged from one to ten inches per second. A section of a film record is shown in Figure 8.

PROCEDURE

Samples of U^{235} , U^{238} , and plutonium were irradiated in the thermal thimble and samples of the same three materials plus U^{238} and thorium were irradiated in the fast thimble. In each case at least four runs were made as follows:

<u>Length of irradiation</u>	<u>Time between end of irradiation and start of count</u>
6 min	100 sec
30 sec	30 sec
9 sec	~ 1.3 sec
3 sec	~ 1.3 sec

The two longer exposures were accomplished from the remote counter position. Timing of irradiation was accomplished by taking the indication of arrival at the irradiation position from the AC bridge mentioned above. The two shorter irradiations were accomplished from the

Figure 5

Delayed Neutron Counting Geometry.

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Figure 6

Delayed Neutron Counting Electronics.

Figure 7

Electronics for delayed neutron counting. Top to bottom on relay rack: audio oscillator, mixer, two high voltage supply - amplifier - scaler combinations, and regulated 110 volt AC supply. At right: Tektronix Oscilloscope Model 511 with Fairchild recording camera.

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Figure 8

Section of film record showing 100 cycle signal and: (1) timing pips separated by 0.1 second, (2) 1000 count pips from Channel A, and (3) 1000 count pips from Channel B. Film speed about 10 inches/second.

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nearer counter position on top of the reactor. The timing was based on the switch interval between energizing (inserting rabbit for irradiation) and de-energizing (withdrawing rabbit) the solenoid valve. The relationship between switch time and actual irradiation time was found by taking a series of runs with varying switch interval. In each run a count was taken during the interval between one and two minutes after the end of irradiation. A plot of the counts against switch interval extrapolated to zero count (Figure 9) gave the intercept representing the difference between switch interval and length of irradiation at constant pressure (7.5 psi). Timing of the switch was manual, a 10-second sweep stop watch being used. Power levels of the reactor were adjusted to give optimum counting rates at the time of interest, i.e., the order of 8,000 - 10,000 counts/second in each channel at the beginning of the recorded count. Power levels were monitored both with the normal reactor instrumentation and with a separate fission counter channel.

ANALYSIS OF DATA

Information of type $N/\Delta t$ was lifted point by point from the film, normalized for number of atoms (of the principal isotope of that sample) and flux ($\propto$ reactor power level) and plotted on semilog paper. Hughes' ³ periods were assumed and a numerical solution⁴ was used on three pairs of arbitrary points of the 30-second irradiation and 6-minute irradiation runs to obtain the saturated counting rates (Y_0^I , Y_0^{II}) at 0 time for the two longest periods. These two counting rates, corrected for shorter irradiation times [$Y_0^I(t) = Y_0^I(1 - e^{-t/\lambda I})$, etc.], were then plotted exponentially on the 9- and 3-second data curves. The usual curve "peeling" procedure was used to obtain $Y_0^{III}(t)$ and $Y_0^{IV}(t)$, which were then converted to saturation irradiation giving Y_0^{III} and Y_0^{IV} . The values given for Y_0^{III} and Y_0^{IV} were the averages of the two values obtained from the 9- and 3-second runs. The results of independent analyses by two different analysts are tabulated in Table II together with the averages. Each analyst's value was his average for both channels.

³D. J. Hughes, et.al., Phys. Rev. 73, 111 (1948).

⁴Assuming the pair of points being used are the counting rate ($Y_{80}(30)$) at 80 seconds on the decay curve of the 30-second irradiation and the counting rate (Y_{200}) at 200 seconds on the decay curve of the saturated irradiation, the following simultaneous equations were solved for Y_0^I and Y_0^{II} :

$$Y_{80}(30) = (1 - e^{-30/80.3}) Y_0^I e^{-80/80.3} + (1 - e^{-30/31.7}) Y_0^{II} e^{-80/31.7}$$

$$Y_{200} = Y_0^I e^{-200/80.3} + Y_0^{II} e^{-200/31.7}$$

Figure 9

Plot to Determine Difference Between Switch
Time and Irradiation Time.

Table II

EXTRAPOLATED COUNTING RATES

	Thermal U ²³⁵	%	Fast U ²³⁵	%	Thermal Pu ²³⁹	%	Fast Pu ²³⁹	%	Thermal U ²³⁵	%	Fast U ²³⁵	%	Fast U ²³⁵	%	Fast Thorium	%
Y _O ^I (55.6)	16,609		7,706		10,156		3,981		17,156		13,077		458		434	
	16,721		7,656		10,114		3,973		17,065		13,038		456		431	
	16,665±13%	5.0	7,682±13%	4.6	10,135±13%	4.2	3,977±13%	4.6	17,111±13%	10.8	13,057±13%	11.2	457±13%	2.0	432±13%	5.7
Y _O ^{II} (22.6)	82,707		41,467		80,297		27,503		46,881		32,091		4,110		1,232	
	81,450		41,899		80,173		27,639		47,036		32,191		4,131		1,225	
	82,078±8%	24.7	41,683±8%	25.2	80,235±8%	33.3	27,571±8%	31.9	46,958±8%	29.7	32,141±8%	27.6	4,120±8%	18.1	1,228±8%	16.2
Y _O ^{III} (4.5)	130,113		72,553		83,439		27,444		66,765		49,853		7,038		2,872	
	131,452		73,447		91,445		31,711		68,799		49,519		7,662		3,085	
	130,782±17%	39.4	73,000±11%	44.2	87,442±11%	36.3	29,577±11%	34.3	67,782±11%	42.9	49,686±11%	42.7	7,350±11%	32.3	2,978±11%	39.2
Y _O ^{IV} (1.5)	108,053		42,889		69,506		27,075		28,857		22,519		10,908		3,248	
	96,219		42,622		56,785		23,061		23,721		20,450		10,707		2,680	
	102,136±25%	30.8	42,755±17%	25.9	63,146±17%	26.2	25,068±17%	29.1	26,289±17%	16.6	21,484±17%	18.5	10,808±17%	47.5	2,964±17%	39.0
Y _O ^T	337,482		164,615		243,398		86,025		159,659		117,541		22,514		7,786	
	325,842		165,624		238,601		86,384		156,621		115,198		22,956		7,421	
	331,662±10%		165,121±5%		240,958±5%		86,193±5%		158,140±5%		116,369±5%		22,735±5%		7,604±5%	

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RELATIVE DELAYED NEUTRON YIELDS

The numbers contained in Table II are merely relative, since reactor flux, spectrum, cross section, and counter efficiency are involved. In order to obtain delayed neutron yields relative to the yield of U^{235} in the slow thimble, we can write, for example:

$$Y_0^T (Pu^{239} \text{ fast}) = A \cdot \text{fission rate}_{Pu^{239}} (\text{in fission/atom/sec})$$

$$\cdot D_{Pu^{239} \text{ fast}}$$

$$Y_0^T (U^{235} \text{ slow}) = A \cdot \text{fission rate}_{U^{235}} \cdot D_{U^{235} \text{ slow}}$$

where

$Y_0^T = Y_0^I + Y_0^{II} + Y_0^{III} + Y_0^{IV}$, A is the efficiency of the system, and D is the delayed neutrons per fission considering, in this case, only the four longest periods. ($D_{U^{235} \text{ slow}} = 0.016$ delayed neutron/fission³.)

Dividing the first equation by the second and transposing, gives

$$\frac{D_{Pu^{239} \text{ fast}}}{D_{U^{235} \text{ slow}}} = \frac{Y_0^T (Pu^{239} \text{ fast})}{Y_0^T (U^{235} \text{ slow})} \cdot \frac{\text{fission rate } U^{235} \text{ slow}}{\text{fission rate } Pu^{239} \text{ fast}}$$

The ratios of the fission rates for all conditions of these irradiations can be obtained by the method developed by Curtis⁵ and used by Kato and Kaufmann.⁶ For Pu^{239} and U^{235} , fission counters activated with the material in question were compared with a U^{235} counter, first in a truly thermal flux (not the thermal thimble) and then in the fast thimble, giving the following relationships:

$$\text{In the thermal flux. } \frac{(CR_{Pu^{239}})}{(CR_{U^{235}})_{\text{thermal}}} = \frac{E_{Pu^{239}} N_{Pu^{239}} \phi_{th} (\sigma_{Pu^{239}th})}{E_{U^{235}} N_{U^{235}} \phi_{th} (\sigma_{U^{235}th})}$$

$$\text{In the fast flux. } \frac{(CR_{Pu^{239}})}{(CR_{U^{235}})_{\text{fast}}} = \frac{E_{Pu^{239}} N_{Pu^{239}} (\phi_{\text{fast}} \sigma_{Pu^{239} \text{ fast}})}{E_{U^{235}} N_{U^{235}} (\phi_{\text{fast}} \sigma_{U^{235} \text{ fast}})}$$

where

CR = counting rate of the counter specified

N = number of atoms of active material in that counter

E = efficiency of the counter.

⁵Departmental Memo.

⁶Reactor Engineering Division Quarterly Report, ANL-5371
(January 15, 1955) pp. 118 - 132.

Dividing the second equation by the first and transposing yields

$$\frac{\frac{(CR_{Pu^{239}})}{(CR_{U^{235}})_{fast}} \cdot \frac{(\sigma_{Pu^{239}th})}{(\sigma_{U^{235}th})}}{\frac{(CR_{Pu^{239}})}{(CR_{U^{235}})_{thermal}}} = \frac{\sigma_{Pu^{239}fast} \phi_{fast}}{\sigma_{U^{235}fast} \phi_{fast}} = \frac{(fission\ rate\ Pu^{239})}{(fission\ rate\ U^{235})_{fast\ thimble.}}$$

where the thermal cross sections can be obtained from BNL-250.⁷

The fission rates of U^{238} and Th were similarly found using, respectively, a counter of natural uranium ($1/140\ U^{235}$) and a counter of Th with an admixture of U^{235} . For example, the fission ratio of U^{238} was obtained as follows:

$$\text{In the thermal flux } \frac{(CR_N)}{(CR_{U^{235}})_{thermal}} = \frac{E_N}{E_{U^{235}}} \frac{1/140\ N_N \sigma_{U^{235}th} \phi_{th}}{N_{U^{235}} \sigma_{U^{235}th} \phi_{th}}$$

since only the U^{235} fraction of the natural U counter will fission in a thermal flux.

In the fast flux

$$\frac{(CR_N)}{(CR_{U^{235}})} = \frac{E_N (1/140\ N_N \sigma_{U^{235}fast} \phi_{fast} + N_N \sigma_{U^{238}fast} \phi_{fast})}{(E_{U^{235}} N \sigma_{U^{235}fast} \phi_{fast})}$$

Dividing the second equation by the first and transposing yields

$$\frac{\frac{(CR_N)}{(CR_{U^{235}})_{fast}}}{\frac{(CR_N)}{(CR_{U^{235}})_{thermal}}} - 1 = 140 \frac{\sigma_{U^{238}fast} \phi_{fast}}{\sigma_{U^{235}fast} \phi_{fast}} = 140 \frac{fission\ rate\ U^{238}fast}{fission\ rate\ U^{235}fast}$$

Ratios of fission rates in the fast and thermal thimbles were taken for U^{235} , U^{238} and Pu by use of a single counter for each material, taking a count first in one thimble and then the other. Figure 10 shows the two types

⁷M. D. Goldberg, D. J. Hughes, J. A. Harvey, V. E. Pilcher, Neutron Cross Sections, Brookhaven National Laboratory, BNL-250 (August 1, 1954)

3	4	5	6	7	8	9	10									
07	22	12	07	61	81	21	91	21	41	81	6	8	2	9	5	4

Figure 10

Fission counters used to measure fission ratios. At top: a "standard" size counter. Center: miniature counter in adapter. Bottom: adapter disassembled to expose miniature counter.

of counters⁸ used in taking these ratios; electronics used were a conventional high voltage supply, pre-amplifier, and scaler.

It had been planned to relate all delayed neutron yields to the yield of U^{235} in a thermal flux; however, the runs made on U^{235} in the thermal thimble were somewhat dubious due to noise and a poor film record, so the results are tabulated (Table III) with reference to the fast fission of U^{235} .

DISCUSSION

Any estimate of the accuracy of these results must consider the subjectiveness of the type of analysis applied to composite counting rate curves. In Table II this is apparent in the difference in results obtained by the analysts; for individual period yields, they differ by as much as 20%. However, the nature of the curve peeling process generally results in errors of alternating sign; that is, if one period is erroneously estimated too high, when it is subtracted, the following period will be too low. This is apparent in that the total yields show a difference between the two analyses of no more than 3%, except in the case of thorium. The discrepancy in per cent yield Y_0^{III} and Y_0^{IV} (Table II) for fast and thermal fission of U^{235} as seen by both analysts is large and probably due to the poor record obtained for some of the runs on the thermal fission of U^{235} , as previously mentioned.

Aside from the physical impossibility of getting close enough to the reactor (i.e., of obtaining short enough transit times) to see the 0.43-second period, the major shortcoming was the lack of knowledge as to the energy response of the delayed neutron counter. No array of neutron sources was available for checking this point. The counting geometry was so arranged that the most direct neutron had to traverse 6 inches of lead and 1-1/2 inches of graphite before reaching a counter. It appears that there must be an appreciable, but unknown, energy effect in response of the counting geometry; there is estimated a factor of 2-1/2 between the highest and lowest delayed neutron energies,⁹ and the minimum of 1-1/2 inches of graphite is insufficient to smooth out the energy differences. This error was arbitrarily estimated on the basis of deviation from a median energy value for yields of individual periods. The error in peeling periods was estimated on the basis of the number of operations performed in obtaining the extrapolated value. Estimates of the errors are tabulated in Table IV.

⁸Reactor Engineering Division Quarterly Report, ANL-5371
(January 15, 1955) pp. 132 - 134.

⁹Reactor Handbook, Vol. I, p. 118.

Table III

DELAYED NEUTRON YIELDS RELATIVE TO U^{235} FAST

Material	Flux	$Y_O^{T(a)}$	Fission Rate	$D^{(b)}$	$Y_O^{T* (c)}$	$D^{(d)}$
			Fission Rate (U^{235} fast)	$D_{U^{235} \text{ fast}}$		$D_{U^{235} \text{ fast}}$
U^{235}	Fast	$165,121 \pm 5\%$	1.00	1.00	$163,853 \pm 5\%$	1.00
U^{235}	Thermal	$331,662 \pm 10\%$	$1.99 \pm 1.5\%$	$1.009 \pm 11\%$	$331,662 \pm 10\%$	$1.017 \pm 11\%$
Pu	Fast	$86,193 \pm 5\%$	$1.30 \pm 3\%$	$.402 \pm 7.5\%$	$86,193 \pm 5\%^{(e)}$	$.405 \pm 7.5\%$
Pu	Thermal	$240,958 \pm 5\%$	$4.09 \pm 3\%$	$.357 \pm 7.5\%$	$240,958 \pm 5\%^{(e)}$	$.360 \pm 7.5\%$
U^{233}	Fast	$116,369 \pm 5\%$	$1.71 \pm 3\%$	$.412 \pm 7.5\%$	$116,011 \pm 5\%$	$.414 \pm 7.5\%$
U^{233}	Thermal	$158,140 \pm 5\%$	$2.44 \pm 3\%$	$.393 \pm 7.5\%$	$158,140 \pm 5\%$	$.396 \pm 7.5\%$
U^{238}	Fast	$22,735 \pm 5\%$	$.062 \pm 2\%$	$2.22 \pm 7.5\%$	$22,680 \pm 5\%$	$2.23 \pm 7.5\%$
Th	Fast	$7,604 \pm 5\%$	$.015 \pm 15\%$	$3.07 \pm 17\%$	$7,604 \pm 5\%$	$3.09 \pm 17\%$

(a) Preliminary 0 time counting rate considering longest four delayed neutron periods (normalized to power level and to number of atoms of principal isotope).

(b) Preliminary delayed neutron yield relative to U^{235} fast.

(c) 0 time counting rate corrected for contribution of principal isotopic impurities.

$$Y_O^{T*} = Y_O^T \left[1 - \frac{\text{fraction of principal impurity} \times \text{relative fission rate of principal impurity} \times \text{relative delayed neutron yield of principal impurity}}{\text{relative fission rate of principal impurity} \times \text{relative delayed neutron yield of principal impurity}} \right]$$

(d) Final result for delayed neutron yield relative to U^{235} fast.

(e) No correction made on Pu due to lack of information about Pu^{240} . See discussion.

Table IV

ESTIMATES OF PROBABLE ERRORS IN EXTRAPOLATED COUNTING RATES

	<u>Power Level</u>	<u>Time of Irradiation</u>	<u>Weight of Sample</u>	<u>Self- Shielding</u>	<u>Counting Losses</u>	<u>Counting Statistics</u>	<u>Peeling Error for Individual Periods</u>	<u>Energy Response</u>	<u>Total</u>
Y_0^I	1%	Negl.	Negl.	Negl.	Negl.	2%	3%	12%	13%
Y_0^{II}	1%	Negl.	Negl.	Negl.	Negl.	2%	3%	7%	8%
Y_0^{III}	1%	2%	Negl.	Negl.	2%	1%	7%	7%	11%
Y_0^{IV}	1%	2%	Negl.	Negl.	2%	1%	12%	12%	17%
Y_0^T	1%	1%	Negl.	Negl.	1%	1%	3%	4%	5%

On the other hand, since all the materials except U^{238} have roughly the same distribution of periods, the error in Y_0^T due to energy response very nearly canceled when compared with another Y_0^T . Accordingly, the error in Y_0^T was estimated at 4% due to lack of knowledge as to energy response and 3% due to plotting except in the case of thermal fission of U^{235} where the film record was not satisfactory.

The errors estimated for the fission ratios in Table V are based on the individual statistics of the data taken plus a certain pessimism based on:

1. The inherent noisiness of the site.
2. The fact that the reactor had previously been run at sustained high power giving an intense gamma background in the fast thimble.

Table V indicates the counting rates on which each ratio is based. The ratio $\frac{\text{fission rate } U^{235} \text{ slow}}{\text{fission rate } U^{235} \text{ fast}}$ depends on only two counting rates using a single counter. The ratios $\frac{\text{fission rate } U^{233} \text{ fast}}{\text{fission rate } U^{235} \text{ fast}}$ and $\frac{\text{fission rate } Pu^{239} \text{ fast}}{\text{fission rate } U^{235} \text{ fast}}$ depend on the taking of four different counting rates (see foregoing equations) instead of the two counting rates necessary for the first ratio cited. To the statistical error must be added the uncertainty in cross section as obtained from BNL-250 ($\sim 1\%$ for U^{235} , $\sim 2\%$ for U^{233} and Pu). To obtain the thermal fission ratios of U^{233} and Pu it was necessary to multiply the two preceding ratios by single counter ratios (i.e., $\frac{\text{fission rate } U^{233} \text{ fast}}{\text{fission rate } U^{235} \text{ fast}} \cdot \frac{\text{fission rate } U^{233} \text{ slow}}{\text{fission rate } U^{233} \text{ fast}} = \frac{\text{fission rate } U^{233} \text{ slow}}{\text{fission rate } U^{235} \text{ fast}}$).

The ratio $\frac{(\text{fission rate } U^{238})}{(\text{fission rate } U^{235})} \text{ fast}$ was obtained as described from four counting rates. However, there is no uncertainty as to thermal cross section; the statistical uncertainty is the only one to be considered.

The counters loaded with thorium plus a "spike" of U^{235} had considerable uncertainty as to the actual fraction of U^{235} ; the active material had been accurately aliquoted, but there had evidently been fractionation in the plating process. Measurements of the fission ratio were not reproducible, ranging from 0.0096 to 0.024. The value cited has been estimated both from the measurements and from theoretical calculations¹⁰ which indicate that

¹⁰H. I. Kraig, private communication.

Table V

ESTIMATES OF PROBABLE ERRORS IN FISSION RATIOS

Fission Ratio	Cross Section	CR _{thermal flux}	CR _{fast thimble}	CR _{thermal thimble}	Total Probable Error in Fission Ratio
$\frac{\text{Pu fast}}{\text{U}^{235} \text{ fast}}$	2% Pu 1% U ²³⁵	1% 1%	1% 1%		} 3%
$\frac{\text{U}^{233} \text{ fast}}{\text{U}^{235} \text{ fast}}$	2% U ²³³ 1% U ²³⁵	1% 1%	1% 1%		} 3%
$\frac{\text{U}^{238} \text{ fast}}{\text{U}^{235} \text{ fast}}$		1% 1%	1% 1%		} 2%
$\frac{\text{Th fast}}{\text{U}^{235} \text{ fast}}$		See Discussion			} 15%
$\frac{\text{U}^{235} \text{ slow}}{\text{U}^{235} \text{ fast}}$			1%	1%	} 1.5%
$\frac{\text{U}^{233} \text{ slow}}{\text{U}^{235} \text{ fast}}$	2% U ²³³ 1% U ²³⁵	1% 1%	1% 1%	1%	} 3%
$\frac{\text{Pu slow}}{\text{U}^{235} \text{ fast}}$	2% Pu 1% U ²³⁵	1% 1%	1% 1%	1%	} 3%
$\frac{\text{Pu slow}}{\text{Pu fast}}$			1%	1%	} 1.5%
$\frac{\text{U}^{233} \text{ slow}}{\text{U}^{233} \text{ fast}}$			1%	1%	} 1.5%

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the $\frac{\text{fission rate Th}}{\text{fission rate U}^{238}}$ which has been measured¹¹ as 0.265 at the center of EBR, decreases approximately 10% in going to the position of the fast thimble used in this experiment. It should be noted that the errors quoted are the "probable errors," i.e., have a 50% chance of being exceeded.

It is interesting to compare the yield of delayed neutrons from thermal and fast fission of U^{233} and Pu. The error due to energy response of the delayed neutron counter effectively drops out; the fission ratio is the ratio of only two counting rates instead of a combination of four or five as when materials are interrelated; and there is no cross section uncertainty in the fission ratio. As a result of these comparisons:

$$\frac{\text{Delayed neutron yield for thermal fission}}{\text{Delayed neutron yield for fast fission}} = 0.956 \pm 6\% \text{ for } \text{U}^{233}$$

$$\frac{\text{Delayed neutron yield for thermal fission}}{\text{Delayed neutron yield for fast fission}} = 0.888 \pm 6\% \text{ for Pu}$$

The second ratio seems to exhibit a real divergence from unity. This appears reasonable in view of the 5% Pu^{240} content in the sample. Pu^{240} has probably a negligible fission cross section in a thermal flux⁷ but Okrent¹² has estimated that the cross section for Pu^{240} in the fast thimble might be of the order of 0.4 that of Pu^{239} . On this basis the 5% fraction of Pu^{240} would have $\sim 2\%$ of the total fissions in the fast thimble. However, should Pu^{240} have a high delayed neutron yield similar to that of U^{238} and Th (likewise even-even nuclei) with 6-9 times the delayed neutron yield of Pu^{239} , this particular sample might exhibit a 12%-18% increase in going from the thermal to the fast thimble, assuming no difference in yield between thermal and fast fission of Pu^{239} . Additional work is needed to determine the delayed neutron yield of Pu^{240} so that its effect on the physics of a Pu- U^{238} breeder or any fast reactor using "dirty" Pu can be assessed.

¹¹Reactor Engineering Division Quarterly Report, ANL-5371
(January 15, 1955) pp. 132.

¹²D. Okrent, private communication.

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