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A MEASUREMENT OF THE AVERAGE FISSION CROSS SECTIONS OF PU^{240} AND PU^{241} IN THE FAST REACTOR NEUTRON SPECTRUM.

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A MEASUREMENT OF THE AVERAGE FISSION CROSS SECTIONS OF PU^{240} AND PU^{241}

IN THE FAST REACTOR NEUTRON SPECTRUM.

The report to follow describes an experiment done to find the average fission cross section of Pu^{240} for the fast neutron spectrum of the fast reactor. The cross section was inferred from a comparison of the fission counting rates from foils of known Pu^{240} enrichments with the rates from standard foils containing only Pu^{239}. Incidental to the Pu^{240} fission cross section measurement the experiment also yielded a measurement of the average fission cross section for Pu^{241}.

I. The Comparison Chamber

In order to measure fission rates from neutron fluxes which had undergone a minimum of energy degradation, a double fission chamber was designed to be inserted all the way into the 5-W reactor port. Thus, when the chamber was in place the foils to be counted were about 1 3/4" from the edge of the active material.

The cylindrical chamber enclosure was machined from Catalin rod to have an inside diameter of 11/16". Two 1/2" x 3/4" collecting electrodes were painted on the inside surface with Aquadag. Slots milled down the sides of the chamber accommodated a silver-plated brass septum, which was soldered to a cap designed to seat into the end of the chamber. Foils to be counted were mounted one on each side of the septum, facing the collecting electrodes. The entire chamber fitted

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into the end of a length of 7/8" steel pipe equipped with a cap which screwed on the end of the pipe and compressed neoprene gasket to form a vacuum-tight system. Seals at the other end of the pipe, which protruded from the edge of the reactor shielding, brought wires from the collecting electrodes out to two preamplifiers. The chamber was operated with 200 volts potential and at a pressure of one atmosphere of argon. When used with model 500 amplifiers the chamber yielded discriminator bias curves with a rate of rise of around 0.1%/v; the total rise amounted to about 4% for the entire "flat" part of the curve.

II. Weighing of Foils

Two foils, designated as A and B, containing different Pu^{240} enrichments were compared with two standard Pu^{239} foils, S and s. Mass spectrographic analyses (by atomic percentages) for the foils follow:

Pu	A (LA 561)	B (LAMS 995)	S, s (LA 561)
238	-	0.06	-
239	92.76	58.1	99.97
240	6.86	34.0	0.03
241	0.38	6.8	-
242	-	1.0	-

Weighing of foils was done in two ways: 1. Alpha counting, and, 2. Thermal fission counting.

1. The ratio of α-particle counting rate from foil x containing several α-active Pu isotopes to that from a "pure" Pu^{239} foil

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S is:

$$\frac{\alpha(X)}{\alpha(S)} = \frac{No(x)\lambda(48) + N_1(x)\lambda(49) + N_2(x)\lambda(4\text{-}10) + N_4(x)\lambda(4\text{-}12)}{N_1(S)\lambda(49)}$$

where: $N_o(x)$ = no. Pu^{238} atoms in foil x

$N_1(x)$ = " Pu^{239} " " " "

$N_2(x)$ = " Pu^{240} " " " "

$N_4(x)$ = " Pu^{242} " " " "

$N_1(S)$ = " Pu^{239} " " std. foil S.

For foil A referred to standard foil S this gives:

$$\frac{N_1(S)}{N_1(A)} = \frac{\alpha(S)}{\alpha(A)}\left(1 + \frac{N_2(A)}{N_1(A)} \cdot \frac{\lambda(4\text{-}10)}{\lambda(49)} + \frac{N_o(A)}{N_1(A)} \cdot \frac{\lambda(48)}{\lambda(49)}\right)$$

$$= \frac{\alpha(S)}{\alpha(A)}\left(1 + 0.074\ \frac{\lambda(4\text{-}10)}{\lambda(49)} + \frac{N_o(A)}{N_1(A)} \cdot \frac{\lambda(48)}{\lambda(49)}\right) \quad (1).$$

neglecting the very slight α-activity of Pu^{241}. For foil B, referred to standard foil s,

$$\frac{N_1(s)}{N_1(B)} = \frac{\alpha(s)}{\alpha(B)}\left(1 + 0.00103\,\frac{\lambda(48)}{\lambda(49)} + 0.585\frac{\lambda(4\text{-}10)}{\lambda(49)} + 0.017\frac{\lambda(4\text{-}12)}{\lambda(49)}\right) \quad (2).$$

Alpha counting was done in a large double chamber designed by L. Rosen. As a check on the symmetry of the counting apparatus the ratios $\alpha(S)/\alpha(A)$ and $\alpha(s)/\alpha(B)$ were taken at several points along the flat portion of the bias curves. These ratios were found to be constant within counting errors over about eighty percent of the α-particle bias curves. The final value of each ratio is the average of the values found at approximately twenty different bias settings.

Although the mass spectrographic analysis of foil A (LA 561) did not reveal the presence of Pu^{238}, a pulse height analysis of its α-particle spectrum did show that α-particles from Pu^{238} contributed a fair amount to the total count. With the help of a planimetric integration of the α-particle spectrum, a value of 0.032 was found for the last term in Eq. 1.

2. Thermal Fission Counting.

The ratios Th(S)/Th(A) and Th(s)/Th(B) of the thermal fission counting rates are related to the Pu^{239} content of the foils by

$$\frac{N_1(S)}{N_1(A)} = \frac{Th(s)}{Th(A)} \left(1 + 0.074 \frac{\sigma_{th}(4\text{-}10)}{\sigma_{th}(49)} + 0.0041 \frac{\sigma_{th}(4\text{-}11)}{\sigma_{th}(49)} \right) \qquad (3).$$

for foils A and S and

$$\frac{N_1(s)}{N_1(B)} = \frac{Th(s)}{Th(B)} \left(1 + 0.0013 \frac{\sigma_{th}(48)}{\sigma_{th}(49)} + 0.585 \frac{\sigma_{th}(4\text{-}10)}{\sigma_{th}(49)} + 0.117 \frac{\sigma_{th}(4\text{-}11)}{\sigma_{th}(49)} + 0.017 \frac{\sigma_{th}(4\text{-}12)}{\sigma_{th}(49)} \right) \qquad (4).$$

for foils B and s.

The thermal fission and subsequent fast fission counting ratios were again determined as an average taken at a number of different bias settings. A further check on the symmetry of the counting apparatus was provided by interchanging the model 500 amplifiers and their associated scalers. This interchange produced observed differences in the ratios which were small in comparison with the counting errors. Counting at a given bias setting was done simultaneously for both sides of the chamber in order to eliminate the effects of small time variations in the reactor level.

It is necessary to choose values for σ_{th} for Pu^{238}, Pu^{239}, and Pu^{242} in Eqs. 3 and 4. Of these a value of 15 b is known for Pu^{238} and less than 50 b is indicated for Pu^{240}(LAMS 995). If the upper limit of 50 b is chosen, the contribution of the Pu^{240} term is still small, and so it together with the Pu^{238} and Pu^{242} terms is neglected. A value of $\sigma_{th}(4\text{-}11) = 1350$ b $= 1.88\ \sigma_{th}(49)$ is taken (LAMS 995).

The values of $N_1(S)/N_1(A)$ and $N_1(s)/N_1(B)$ are critically dependent on the decay constants in Eqs. 1 and 2, and although some spread still exists in the reported measurement of the Pu^{240} half-life, most of these have been around 6500 y. In the table which follows several values of $T_{1/2}$ (4-10) have been selected to indicate their influence on the self-consistency of the counting data. In each case the values $T_{1/2}$ (49) $= 24.2 \times 10^3$ y (LAMS 995) and $T_{1/2}$ (48) $= 89.6$ y (ANL 4405) were used. The term in Eq. 2 due to Pu^{242} was neglected in view of its relatively low abundance and its predicted slow decay ((H) CF-3211).

	$N_1(S)/N_1(A)$		$N_1(s)/N_1(B)$	
$T_{1/2}$ (4-10)	α-ctg	Th fission	α-ctg	Th fission
6000 y.	1.145 ± .0018	1.123 ± .0017	5.02 ± .0055	4.88 ± .005
6250 y.	1.133 ± .0018	"	4.89 ± .0055	"
6500 y.	1.123 ± .0018	"	4.77 ± .0055	"

The validity of assuming a negligible effect from $\sigma_{th}(4\text{-}10)$ is supported by the fact that any increase in $N_1(S)/N_1(A)$ or $N_1(s)/N_1(B)$

as found by thermal fission counting would destroy the agreement with those values which were determined by alpha-counting.

The half-life figure arrived at from weighing foil B is probably the more reliable one since the relative contribution from Pu^{240} was higher than it was in foil A.

III. Fast Neutron Cross Sections

From the ratios F(S)/F(A) and F(s)/F(B), found by fission counting in the fast neutron flux, one gets the relation

$$\frac{\bar{\sigma}(4\text{-}10)}{\bar{\sigma}(49)} = \frac{1}{0.074}\left(\frac{F(A)}{F(S)} \cdot \frac{N_1(S)}{N_1(A)} - 1 - 0.0041\,\frac{\bar{\sigma}(4\text{-}11)}{\bar{\sigma}(49)}\right)$$

for foils A and S, and

$$\frac{\bar{\sigma}(4\text{-}10)}{\bar{\sigma}(49)} = \frac{1}{0.585}\left(\frac{F(B)}{F(s)} \cdot \frac{N_1(s)}{N_1(B)} - 1 - 0.001\,\frac{\sigma(48)}{\sigma(49)} - 0.117\,\frac{\bar{\sigma}(4\text{-}11)}{\bar{\sigma}(49)} - 0.017\,\frac{\bar{\sigma}(4\text{-}12)}{\bar{\sigma}(49)}\right)$$

for foils B and s, where the $\bar{\sigma}$'s refer to the average fission cross section for the reactor fast neutrons. Weighing by thermal fission counting is taken to give the more reliable value for $N_1(S)/N_1(A)$. The two equations above yield

$$\frac{\bar{\sigma}(4\text{-}10)}{\bar{\sigma}(49)} = \frac{1}{.074}\left(0.915 \times 1.126 - 1 - .0041\,\frac{\bar{\sigma}(4\text{-}11)}{\bar{\sigma}(49)}\right)$$

$$= \frac{1}{.584}\left(0.282 \times 4.88 - 1 - 0.117\,\frac{\bar{\sigma}(4\text{-}11)}{\bar{\sigma}(49)}\right)$$

where contributions from Pu^{238} and Pu^{242} fast neutron fission have been neglected. Solving these for $\bar{\sigma}(4\text{-}10)/\bar{\sigma}(49)$ and $\bar{\sigma}(4\text{-}11)/\bar{\sigma}(49)$ gives

$$\bar{\sigma}(4\text{-}11)/\bar{\sigma}(49) = 1.62 \pm 0.26$$

$$\bar{\sigma}(4\text{-}10)/\bar{\sigma}(49) = 0.32 \pm 0.052$$

The indicated limits of error arise from the r.m.s. counting errors only and are therefore probably optimistic. During the course of fission counting it was found that interchanging the two foils in the chamber caused differences in the ratios which were somewhat outside the counting errors. An arithmetic mean was taken as the proper value of the ratio; if the observed difference arises, say, from a deficiency of counting solid angle on one side of the chamber, then the average ratio will differ from the true ratio by an amount which is of the order of the square of the fractional deficiency.

The value of $\bar{\sigma}(4\text{-}10)/\bar{\sigma}(49)$ reported here is in fair agreement with a measurement of $(\nu-1-\alpha)\,\bar{\sigma}(4\text{-}10)/(\nu-1-\alpha)\,\bar{\sigma}(49) = 0.4 \pm 0.1$ made by D. and J. Hall by the method of reactor "danger coefficients" (P-Div. Monthly Report, January, 1949).

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