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HIGH-ENERGY MEASUREMENTS OF NEUTRON-INDUCED
FISSION CROSS-SECTION RATIOS
INVOLVING ^{233}U , ^{235}U , ^{238}U , AND ^{239}Pu
USING THE METHOD OF THRESHOLD CROSS SECTIONS

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ABSTRACT

Fission cross section measurements for a wide collection of uranium and plutonium isotopes are being conducted at LLL using the 100-MeV electron linear accelerator (Linac) as a pulsed neutron source. The cross section ratios are measured as a function of neutron energy from 1 keV to 30 MeV using the time-of-flight technique.

To obtain fission cross section ratios above 1 MeV, we used the threshold cross section method.¹ This method allows us to obtain the ratios without depending on normalizations to absolute cross section measurements. Knowledge of neither the amount of fissionable mass nor the efficiency for detecting fissions in the fission chambers is required for the analysis. This advantage eliminates a potentially large uncertainty in our ratio measurements.

Preliminary results for the fission cross section ratios $^{238}\text{U}/^{233}\text{U}$, $^{238}\text{U}/^{235}\text{U}$, and $^{238}\text{U}/^{239}\text{Pu}$ extend from 1 to 30 MeV with an energy resolution of at least 5% and counting uncertainties less than 5% over most of the energy range. Additional preliminary measurements of the fission cross section ratios $^{233}\text{U}/^{235}\text{U}$ from 1 keV to 30 MeV, $^{238}\text{U}/^{235}\text{U}$ from 750 keV to 30 MeV, and $^{239}\text{Pu}/^{235}\text{U}$ from 10 keV to 30 MeV are reported with an energy resolution of at least 5% and counting uncertainties less than 3% over most of the energy range. These ratios were normalized using the threshold cross section method.

INTRODUCTION

A large number of measurements has been reported in the literature for the past two decades on fission cross section ratios.²⁻²⁸ Figures 1 to 6 summarize the measurements published for the ratios $^{233}\text{U}/^{235}\text{U}$, $^{238}\text{U}/^{235}\text{U}$, and $^{239}\text{Pu}/^{235}\text{U}$. The large discrepancies apparent in the data sets may be attributed in part to normalization uncertainties and to the failure of any single measurement to span the entire energy range of interest. Applying the threshold cross section method to data taken with a neutron energy spectrum that is continuous from 1 keV to more than 30 MeV can eliminate these problems. We have used this method in the analysis of our experiments. The LLL 100-MeV electron linear accelerator provided the desired neutron spectrum. We conducted the experiment at the 34-m station on the 250-m time-of-flight tube. This report describes the experiment and presents the preliminary results of the cross section ratios involving ^{233}U , ^{235}U , ^{238}U , and ^{239}Pu .

(text continued on p. 8)

Fig. 1. Fission cross-section ratio $^{233}\text{U}/^{235}\text{U}$ from publications prior to 1966.

Fig. 2: Fission cross-section ratio $^{233}\text{U}/^{235}\text{U}$ from publications after 1965.

Fig. 3. Fission cross-section ratio $^{238}\text{U}/^{235}\text{U}$ from publications prior to 1966.

Fig. 4. Fission cross-section ratio $^{238}\text{U}/^{235}\text{U}$ from publications after 1965.

Fig. 5. Fission cross-section ratio $^{239}\text{Pu}/^{235}\text{U}$ from publications prior to 1966.

Fig. 6. Fission cross-section ratio $^{239}\text{Pu}/^{235}\text{U}$ from publications after 1965.

THE EXPERIMENT

Threshold Cross Section Method

To measure a fission cross section ratio by the threshold cross section method¹ one uses two fission chambers. The first contains a mixture of the two isotopes of interest with an atom ratio η of the isotope B to the threshold isotope A. For some range of energies below the threshold of isotope A, the ratio of fission cross sections $\sigma_A(E)/\sigma_B(E)$ must be negligible compared with the same ratio above the threshold. The expected count rate in this mixed chamber is

$$r_m(E) = \phi(E) N_A [\sigma_A(E) + \eta \sigma_B(E)] ,$$

where $\phi(E)$ is the neutron flux and N_A is the effective number of atoms of isotope A. The second fission chamber contains an effective number N_B of atoms of pure isotope B. The rate for this pure chamber is

$$r_p(E) = \phi(E) N_B \sigma_B(E) .$$

The two chambers are exposed simultaneously to the same neutron beam to measure $r_m(E)$ and $r_p(E)$. Their ratio gives

$$R(E) = \frac{r_m(E)}{r_p(E)} = \frac{N_A}{N_B} \left(\frac{\sigma_A(E)}{\sigma_B(E)} + \eta \right) .$$

Below the threshold of isotope A, $R(E)$ is a constant that will be called Q , and the experimental results give the ratio of effective numbers of atoms: $N_A/N_B = Q/\eta$. Substituting for N_A/N_B in the equation for $R(E)$ and solving for $\sigma_A(E)/\sigma_B(E)$, one obtains

$$\frac{\sigma_A(E)}{\sigma_B(E)} = \eta \left(\frac{R(E)}{Q} - 1 \right) \quad (1)$$

with no further normalization needed.

In the present experiment we used three composition fission chambers. The atom ratio η was determined using mass spectrometric methods for the $A = {}^{233}\text{U}$, $B = {}^{238}\text{U}$ and the $A = {}^{235}\text{U}$, $B = {}^{238}\text{U}$ composition fission chambers; samples were analyzed by groups at LLL, Los Alamos Scientific Laboratory, and Dow Chemical's Rocky Flats plant. The atom ratio ${}^{233}\text{U}/{}^{238}\text{U}$ was measured to be 0.1453 ± 0.0004 and the ${}^{235}\text{U}/{}^{238}\text{U}$ ratio was 0.3398 ± 0.0008 . For the $A = {}^{239}\text{Pu}$, $B = {}^{238}\text{U}$ chamber the atom ratio was determined at LLL using both coulometric methods and mass spectrometric analysis and was found to be 0.2284 ± 0.0017 ; further analysis is in progress to determine this atom ratio more precisely. Having these composition fission chambers allows us to determine the following ratios as a function of energy without normalizing to other cross section measurements:

- 1) ${}^{238}\text{U}/{}^{233}\text{U}$,
- 2) ${}^{238}\text{U}/{}^{235}\text{U}$,
- 3) ${}^{238}\text{U}/{}^{239}\text{Pu}$,

$$4) \frac{{}^{233}\text{U}}{{}^{235}\text{U}} = \frac{{}^{238}\text{U}/{}^{235}\text{U}}{{}^{238}\text{U}/{}^{233}\text{U}},$$

$$5) \frac{{}^{239}\text{Pu}}{{}^{235}\text{U}} = \frac{{}^{238}\text{U}/{}^{235}\text{U}}{{}^{238}\text{U}/{}^{239}\text{Pu}}.$$

Fission Chambers

Figure 7 shows the basic design of the fission chambers used in our experiments. Four modular fission chambers were connected back-to-back and placed in a pressure vessel. We used a modular design to prevent cross-contamination of the fissionable materials. Each modular chamber contained an odd number of fission foils, either 5 or 7. Table 1 gives the areal densities of fissionable material and a description of the contents of our fission chambers.

Table 1. Description of fission chambers.

Detector No.	Modular fission chamber No.	Description ^a	Total fissionable material (estimated) (mg)	Areal density per coated surface (g/m ²)
1	1	5 foils of ²³⁵ U	135	2.98
	2	7 foils of ²³⁹ Pu	166	2.61
	3	5 foils of ²³⁸ U + ²³⁹ U	128	2.82
	4	5 foils of ²³⁵ U	135	2.98
2	1	5 foils of ²³⁵ U	135	2.98
	2	7 foils of ²³⁸ U	198	3.12
	3	7 foils of ²³⁸ U + ²³⁵ U	243	3.83
	4	5 foils of ²³⁵ U	135	2.98
3	1	5 foils of ²³⁵ U	135	2.98
	2	7 foils of ²³³ U	173	2.72
	3	5 foils of ²³⁸ U + ²³³ U	174	3.84
	4	5 foils of ²³⁵ U	135	2.98

^aEach foil was coated on both sides with fissionable material. Foil coating was restricted to a 76 mm diameter.

The fission foils, made at LLL using a painting technique,^{29,30} contained uranium and plutonium isotopes whose isotopic purities are shown in Table 2. The fissionable material applied to both sides of each foil was confined to a 38 mm radius from the center of the 66-mm-radius aluminum substrates. Two areal densities, 24.0 and 34.3 g/m², of aluminum were used for the substrate material. The collimated

Fig. 7. Fission chamber and its immediate environment. The design chosen for the double containment enables us to place an additional ground connection on the fission chamber. Protection of our fission chambers from neutron back-grounds is obtained by surrounding the fission chamber assembly with a cadmium-lined cylindrical can.

Table 2. Isotopic analysis of high-purity isotopes. Values are in atom percent.

Sample	^{232}U	^{233}U	^{234}U	^{235}U	^{236}U	^{238}U
High-purity ^{233}U	6×10^{-10}	99.9986	0.0003	0.00004	0.0009	0.00008
High-purity ^{235}U	--	$<1 \times 10^{-6}$	0.0297	99.912	0.0166	0.0419
High-purity ^{238}U	--	$<1 \times 10^{-6}$	$<1 \times 10^{-6}$	6×10^{-6}	$<1 \times 10^{-6}$	99.999
Sample	^{238}Pu	^{239}Pu	^{240}Pu	^{241}Pu	^{242}Pu	^{244}Pu
High-purity ^{239}Pu	<0.0002	99.978	0.021	0.0005	0.0005	--

beam radius at the 34-m time-of-flight station was 40 mm excluding penumbra, 52 mm including penumbra. Having the foils coated to within a maximum radius of 38 mm allows us to place the fissionable material within the main neutron beam. The fission chamber pressure containments, also made of aluminum, were designed to produce minimal perturbation of the incident beam. Filled to a pressure of 127 kPa (18.5 psia) with a gas mixture of 90% argon and 10% carbon dioxide, the fission chamber has an operating voltage of 500 V. Plate spacings of 4.0 and 4.5 mm are used.

Because of the high alpha activity of ^{233}U and ^{239}Pu , the fissionable material had to be doubly contained; Fig. 7 shows how we accomplished this. The design chosen for the double containment enables us to place an additional ground connection on the fission chamber. We used triaxial cable for our signal lines; the center wire carried the +500 V bias, the first braid connected to the first containment, and the outer braid connected to the second containment. The double containment requirement was thereby put to good use because it provided us with a double electrical shield for the fission chamber. We found the double shield helpful in eliminating pickup problems for the Linac.

Counting Electronics

Figure 8 is a block diagram of the counting electronics. The amplifiers for the fission chambers were designed by E. Ables of the LLL Electronics Department, who used an outline for designing fast current amplifiers.³¹ With a gain of 2000, an input impedance of 50Ω and a rise time of 3.5 ns, this amplifier design provided excellent suppression of alpha particle pileup when connected to the ^{233}U and ^{239}Pu fission chambers. Typical fission fragment pulses had rise times of 12 ns, full-width half-maximum values of 30 ns, and average fission-fragment-pulse-to-noise ratio of about 5.

The timing resolution of the fission chamber system has been found to be better than the rise time of 12 ns. Measurements indicate that our timing resolution lies between 6 and 9 ns.

Fig. 8. Diagram of counting electronics and data-acquisition system.

Signals from the current amplifiers travel along fast coaxial cables from the detector location to the experimental counting room. There, linear fanouts duplicate each signal; one is sent to a discriminator, the other to a linear gate and stretcher amplifier. Gated by the discriminator, the signals are shaped by the stretcher amplifier and sent on to the pulse-height-analyzer system. Other outputs from the discriminator go to a count-rate meter and the two time digitizers. The count-rate meter monitors the counting rates going into the time digitizers and acts as a safety control. Data are analyzed by a Digital Computer Corp.* D112 computer-based data-acquisition system and/or the PDP-15 system. The PDP-15 system has the added advantage of pulse-height as well as time-of-flight information on each data event.

Data System

Events as a function of neutron time of flight are recorded simultaneously for each of the three or four detectors used in each experimental run. The EG&G time digitizer determines the time interval between Linac start pulses and fission chamber stop pulses. Each time is accompanied by a four-bit tag to identify the detector producing the stop and is sent to the D112 computer through an interface. For each detector, counts vs time-of-flight channel are recorded in the computer, with channel widths increasing in steps from 0.016 to 1.024 μ s; the time of flight spans a total time of about 90 μ s, which corresponds to a neutron energy of 750 eV. A CRT scope displays the counts per channel as a function of channel while the data is being taken so that the operation of the system can be visually monitored. Data analyzed by the D112 are recorded on magnetic tape and/or paper tape. All data processing is done on LLL's CDC 6600 and 7600 computers.

The time digitizer was set to have a 4- μ s dead time and the data was corrected for dead time although this correction was small (less than 2%). It was divided out in the ratio of one chamber to another since all were at essentially the same distance from the neutron source. There was an additional effect similar to dead time due to falsification of detector tags. If an event was followed within 4 μ s by a second event, the tag given to the computer was the logical "OR" of the tags for the detector of the first event and the detector of the second event. This effect was about the same size as the dead time effect and corrections were made to the data.

As a check on the data system and the corrections for dead time and false tags, we made a run using the plutonium fission chambers with no neutron source; we set the bias low enough to count alpha particles at a large rate and used a pulser to provide the starts. We corrected the resulting data for dead time and false tags. After these corrections, it was constant as a function of time to better than 2%.

* Reference to a company or product name does not imply approval or recommendation of the product by the University of California or the U. S. Atomic Energy Commission to the exclusion of others that may be suitable.

Although all the data in this report was taken on the D112 data system, we also took data simultaneously on E Division's PDP-15 system during some runs. The PDP-15 data included a pulse height and time of flight for each event. We observed no discrepancies between data taken on the two systems. This provided a cross check on our timing because it was done independently with a second EG&G digitizer on the PDP-15 system. The performance of our D112 software is also verified by this comparison to data taken with completely independent software. The pulse height information from the PDP-15 aided us in choosing bias levels for our D112 system.

Linac and Time-of-Flight Facility

The LLL electron linear accelerator (Linac) is used to provide a pulsed source of neutrons. With all five sections in use, the accelerator can deliver 115 MeV electrons to the above-ground neutron cell. There the electrons strike a water-cooled tantalum target at which bremsstrahlung occurs. This in turn produces the neutrons by the (γ, n) reaction. For our experiment an electron pulse width of 10 ns and a repetition rate of 1440 Hz was chosen.

Figure 9 shows the 250-m time-of-flight facility. Figure 10 shows a partial view of the experimental detector tank at the 34-m station at which the experiment is conducted. This tank has a diameter of 1.2 m and a length of 2.4 m. Side access ports enable us to install the fission chambers. Figure 11 shows a typical view of the inside of the experimental tank. Protection of our fission chambers from neutron backgrounds is obtained by surrounding the fission chamber assemblies with a cadmium-lined cylindrical can (Fig. 7) containing natural boron carbide. This is then located at the center of the experimental tank, which in turn is surrounded by boric acid blocks. Pressure transducer and signal lines feed to the outside electronics through a bulkhead connection in the experimental tank. Once a fission chamber is inside, the outside of the experimental tank is surrounded with boric acid blocks. Figure 10 also shows a partial view of the thermally insulated electronics house, which contains the amplifiers for our fission chambers. These signals then travel along fast coaxial cables to the counting electronics shown in Fig. 8.

Proper collimation of the neutron beam is achieved using five sets of collimators between the target and the experiment. At the upstream end of the time-of-flight tube is an overlap filter consisting of enriched boron-10 loaded in polyethylene (areal density is about 10 kg/m^2) backed up with 1.5 mm of cadmium.

We determined the neutron backgrounds by conducting comparison experiments with glass and aluminum absorbers. Figures 12-14 give the results of these measurements. Figure 12 shows the measured background at the 2.8-keV sodium resonance, at which the expected transmission of neutrons is e^{-18} . Figure 13 is the 35-keV aluminum resonance, at which the transmission is $e^{-6.6}$. Figure 14 shows the 440-keV oxygen resonance; the expected transmission here is $e^{-5.7}$, whereas the measurement is $e^{-5.5}$.

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Fig. 9. Above-ground portion of the Livermore electron linear accelerator (Linac).

Fig. 10. Experimental detector tank at the 34-m time-of-flight station. The tank has a diameter of 1.2 m and a length of 2.4 m. Side access ports enable us to install the fission chambers. Once the fission chamber is inside, the experimental tank is surrounded with boric acid blocks.

Fig. 11. Inside view of experimental tank. The fission chamber assembly, surrounded by a cadmium-lined cylindrical can containing natural boron carbide, is located at the center of the tank.

Fig. 12. Ratio of count rate with absorber in the beam to rate without absorber in for a uranium-235 fission chamber for neutron energies between 1 and 10 keV. The large dip at 2.85 keV is the effect of the sodium resonance. The out-of-time neutron background is less than 0.01% at this resonance.

Fig. 13. Ratio of count rate with absorber in the beam to rate without absorber in for a uranium-235 fission chamber for neutron energies between 10 and 100 keV. The large dip at 36 keV is due to the aluminum resonance. The out-of-time neutron background is less than 0.01% at this resonance.

Fig. 14. Ratio of count rate with absorber in the beam to rate without absorber in for a uranium-235 fission chamber for neutron energies between 100 and 1000 keV. The dip at 430 keV is due to the oxygen resonance. The expected transmission at that resonance is indicated by the line at 3.3×10^{-3} . The background is less than 0.1% at this energy.

Timing and Resolution

To conduct an experiment using the time-of-flight technique, it is essential to have an accurate time reference. The gamma flash from the tantalum target provided us with an interesting means for determining our timing. The effects of the gamma flash on the fission chamber were greatly reduced by placing a 13-mm lead sheet over the time-of-flight tube opening, just upstream of the overlap filter. High-energy gamma rays, however, still go down the tube and strike our detectors. These photons produce a pulse-height distribution that is very similar to that of the neutron-produced fission pulses occurring at later times. We suspect that the former pulses are due to photo-fission events in the fissionable materials.

Looking at only these pulses, we split the counts in the gamma flash between two time-of-flight channels in the computer. This split is fine-tuned by adding delay cables to either our start or stop lines of the EG&G time digitizer. We verified our timing by looking at the high-energy resonances of carbon.

The resolution for our experiment is determined by the resolution of the fission chamber system (≤ 9 ns) and the pulse width of the electron pulses striking the tantalum target (~ 10 ns). Taken in quadrature, these two components result in a resolution of about 13 ns. Data was taken with a minimum time per channel of 16 ns, which corresponds to 5.4% energy resolution at 20 MeV at our 34.25-m time-of-flight station. The added component to the resolution due to uncertainties in distance, target size, and detector size are small compared with the above two components. Our time to energy conversion includes a relativistic correction.

Efficiencies of the Fission Chambers

Table 3 gives the estimated average efficiencies relative to the uranium-235 chamber of the fission chambers used in these measurements. Pulse height analysis showed good separation between fission fragment and noise signals in the uranium-235 chambers, and their efficiency is estimated to be at least 0.98. For the other chambers, uncertainties in masses of fissile material and in cross sections make efficiency estimates uncertain by as much as 7%.

The areal densities of fissile material for the fission chambers used here range from 2.6 to 3.8 g/m², as shown in Table 1. For these areal densities and an isotropic angular distribution of fission fragments, at least 1 to 2% of the fission events in the chamber are not detected because the fragments stop in the foil. The fraction of undetected events changes with neutron energy as the fragment angular distribution changes with energy. The energy dependence in the efficiency due to anisotropy can be calculated for isotopes whose angular distributions have been measured if all the inefficiency is caused by loss of fragments in the foil.³² These calculations show a very small effect (less than 1%).

When high alpha activity makes a high bias necessary, the efficiency is lowered and fragments other than those stopping in the foil can be rejected. These might

Table 3. Estimated efficiencies of the fission chambers and upper bounds on the effect of fragment angular anisotropy on efficiency.

Chamber	Efficiency	1.75-4.0 MeV interval		(n, n'F) anisotropy peak	
		Anisotropy ^a	% effect	Anisotropy ^a	% effect
²³⁹ Pu	0.74	1.10	1.1	1.22	2.3
²³⁸ U + ²³⁹ Pu	0.89	1.28	1.6	1.87	4.2
²³⁸ U	0.98	1.28	0.3	1.87	0.9
²³⁸ U + ²³⁵ U	0.98	1.28	0.3	1.87	0.9
²³³ U	0.91	1.13	0.6	1.33	1.5
²³⁸ U + ²³³ U	0.94	1.28	0.9	1.87	2.5

^aAnisotropy is defined as the ratio of the angular distribution at 0° to the distribution at 90° relative to the neutron direction.³³

include fragments that travel straight across the gap between chamber plates, depositing less ionization in the chamber than fragments that cross obliquely. This can also result in an anisotropy effect on chamber efficiency.

One can calculate an upper bound to the magnitude of the change in efficiency that would be caused by a change from an isotropic to an anisotropic angular distribution of fission fragments. The calculation requires the original efficiency and the shape of the angular distribution. The maximum effect occurs when all the fragments in one range of angles are lost while all of the fragments at other angles are detected. We calculated the maximum effect of anisotropy on fission chamber efficiencies for the efficiencies of Table 3, using the measured anisotropies of Henkel and Simmons³³ and a two-term Legendre-polynomial angular distribution in each case. The limit on the anisotropy effect was calculated for the 1.75-4.0 MeV normalization interval and for the peak in anisotropy near the (n, n'F) threshold for ²³³U, ²³⁸U, and ²³⁹Pu. Table 3 gives the results for these bounds. They should be viewed only as upper limits to the effect of anisotropy on the efficiencies and not as corrections to the data.

PRELIMINARY DATA

²³⁸U/²³³U, ²³⁸U/²³⁵U, and ²³⁸U/²³⁹Pu Fission Cross-Section Ratios as a Function of Energy

Figures 15-17 show our preliminary data on the fission cross section ratios ²³⁸U/²³³U, ²³⁸U/²³⁵U, and ²³⁸U/²³⁹Pu as a function of energy using the threshold cross section method. The ratios were calculated for individual experimental runs, compared for consistency, and then averaged to obtain the results given here. The error bars in these figures represent the counting errors in the individual points. The uncertainties in these results due to uncertainty in η and Q (see Eq. 1) are not represented in the individual error bars because they are overall uncertainties for

(text continued on p. 26)

Fig. 15. Fission cross-section ratio $^{238}\text{U}/^{233}\text{U}$ between 1 and 30 MeV. At energies (E) below 5 MeV the data were averaged in 4% $\Delta E/E$ intervals.

Fig. 16. Fission cross-section ratio $^{238}\text{U}/^{235}\text{U}$ between 1 and 30 MeV. At energies (E) below 5 MeV the data were averaged in 4% $\Delta E/E$ intervals.

Fig. 17. Fission cross-section ratio $^{238}\text{U}/^{239}\text{Pu}$ between 1 and 30 MeV. At energies (E) below 5 MeV the data were averaged in 4% $\Delta E/E$ intervals.

each set of data points. The average Q values used to obtain the absolute ratios and their uncertainties are:

Ratio	Q
$^{238}\text{U}/^{233}\text{U}$	0.119469 ± 0.000570
$^{238}\text{U}/^{235}\text{U}$	0.482008 ± 0.001059
$^{238}\text{U}/^{239}\text{Pu}$	0.177905 ± 0.000824

These Q values were obtained from the ratios of the sums of counts in the composite chambers and pure isotope chambers. The counts were summed over energy intervals chosen to avoid significant fission cross sections for ^{238}U , scattering corrections for the aluminum of the chambers, and large no-beam background corrections.

Assuming the same efficiency for each fission chamber, estimates of these Q values can be made by using the values of η and the total masses of fissionable material given in Table 1. The estimated Q values are:

Ratio	Estimated Q
$^{238}\text{U}/^{233}\text{U}$	0.13 ± 0.009
$^{238}\text{U}/^{235}\text{U}$	0.46 ± 0.032
$^{238}\text{U}/^{239}\text{Pu}$	0.14 ± 0.010

An estimated uncertainty of $\pm 7\%$ is assigned to these Q values; this uncertainty arises from the accuracy of the values in Table 1.

$^{233}\text{U}/^{235}\text{U}$, $^{238}\text{U}/^{235}\text{U}$, and $^{239}\text{Pu}/^{235}\text{U}$ Fission Cross-Section Ratios as a Function of Energy

Figures 18-23 show our preliminary data on the ratios $^{233}\text{U}/^{235}\text{U}$, $^{238}\text{U}/^{235}\text{U}$, and $^{239}\text{Pu}/^{235}\text{U}$. Our ratios were normalized to numbers obtained from the ratios reported on p. 8; we chose the 1.75-4.00 MeV energy interval as the normalization interval. The averages from 1.75 to 4 MeV of the ratios were formed by weighting the values of the individual ratios in their 16-ns time-of-flight channels by the energy width of the channels as follows:

$$\bar{a} = \frac{1}{\sum_i \Delta E_i} \left[\sum_i a_i \Delta E_i \right]. \quad (2)$$

To calculate the uncertainty in $\bar{a}$ as defined above, one uses the relation $a_i = \eta(R_i/Q - 1)$, as discussed in the section on the threshold cross section method. The result when substituting for a_i in Eq. 2 is:

$$\bar{a} = \eta(\bar{R}/Q - 1),$$

(text continued on p. 33)

Fig. 18. Fission cross-section ratio $^{233}\text{U}/^{235}\text{U}$ between 0.1 and 30 MeV. At energies (E) below 5 MeV the data were averaged in 4% $\Delta E/E$ intervals.

Fig. 19. Fission cross-section ratio $^{233}\text{U}/^{235}\text{U}$ between 1 and 100 keV. The data are averaged in 10% $\Delta E/E$ energy intervals.

Fig. 20. Fission cross-section ratio $^{238}\text{U}/^{235}\text{U}$ between 1 and 30 MeV. At energies (E) below 5 MeV the data were averaged in 4% $\Delta E/E$ intervals.

Fig. 21. Fission cross section ratio $^{238}\text{U}/^{235}\text{U}$ between 0.75 and 2.0 MeV.

Fig. 22. Fission cross-section ratio $^{239}\text{Pu}/^{235}\text{U}$ between 0.1 and 30 MeV. At energies (E) below 5 MeV the data are averaged in 4% $\Delta E/E$ intervals.

Fig. 23. Fission cross-section ratio $^{239}\text{Pu}/^{235}\text{U}$ between 10 and 100 keV. The data are averaged in 10% $\Delta E/E$ energy intervals.

with

$$\bar{R} = \frac{1}{\sum_i \Delta E_i} \left[\sum_i R_i \Delta E_i \right].$$

The uncertainty in $\bar{a}$ due to the statistical uncertainties of η , Q , and the R_i is:

$$\delta \bar{a} = \left[\left(\delta \eta (\bar{R}/Q - 1) \right)^2 + \left(\delta Q \frac{n \bar{R}}{Q^2} \right)^2 + \left(\frac{\eta}{Q \sum_i \Delta E_i} \right)^2 \sum_i (\delta R_i \Delta E_i)^2 \right]^{1/2}.$$

We determined the following values for the cross section ratios:

- 1) $^{238}\text{U}/^{233}\text{U}$ (1.75-4.00 MeV) = 0.3023 ± 0.0026
- 2) $^{238}\text{U}/^{235}\text{U}$ (1.75-4.00 MeV) = 0.4404 ± 0.0035
- 3) $^{238}\text{U}/^{239}\text{Pu}$ (1.75-4.00 MeV) = 0.2760 ± 0.0044 .

Using these numbers we can then determine the following for the interval 1.75-4.00 MeV:

- 4) $^{233}\text{U}/^{235}\text{U} = \frac{^{238}\text{U}/^{235}\text{U}}{^{238}\text{U}/^{233}\text{U}} = 1.457 \pm 0.017$
- 5) $^{239}\text{Pu}/^{238}\text{U} = \frac{^{238}\text{U}/^{235}\text{U}}{^{238}\text{U}/^{239}\text{Pu}} = 1.596 \pm 0.028$.

These are the numbers used to normalize the ratios shown in Figs. 18-23.

SUMMARY

Figures 15 to 23 give our preliminary results for neutron-induced fission cross section ratios involving ^{233}U , ^{235}U , ^{238}U , and ^{239}Pu using the threshold cross section method. These results confirm the expected advantage of the method in obtaining cross section ratios whose normalization depends only on the measurement of the atom ratio for the composition fission chambers. Further ratio measurements for these isotopes and some other isotopes are in progress. In future measurements we plan to store both the pulse height and the time of flight information during data acquisition.

These preliminary results may be changed as a result of further analysis of systematic effects and better statistics in the measurements of the same ratios. Rough estimates of the scattering of neutrons in the aluminum of the chambers and foils show it to be a negligible effect except possibly at resonance peaks. A more detailed analysis of scattering is being planned. We are continuing to try to obtain cross checks and improvements in the accuracy of the atom ratios η for the composition chambers. More detailed pulse height analysis will help determine if the use of a constant bias level introduces energy dependence in the efficiency for detecting fission events. Modification of our counting electronics will eliminate or greatly reduce the correction for false detector tags.

When finalized, our cross-section-ratio data will be released in tabular form.

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REFERENCES

1. J. W. Behrens, Determination of Absolute Fission Cross Section Ratios Using the Method of Threshold Cross Sections, Lawrence Livermore Laboratory, Rept. UCRL-51478 (1973).
2. R. L. Henkel, Fission Excitation Curves for U^{233} , U^{235} , U^{236} , U^{238} , Np^{237} , Los Alamos Scientific Laboratory, N. Mex., Rept. LA-1495 (1952).
3. R. W. Lamphere, "Fission Cross Sections of the Uranium Isotopes, 233, 234, 236 and 238, for Fast Neutrons, " Phys. Rev. **104**, 1654 (1956).
4. R. L. Henkel, A Summary of Neutron-Induced Fission Cross Sections, Los Alamos Scientific Laboratory, N. Mex., Rept. LA-2114 (1957).
5. R. L. Henkel, Fast Neutron Cross Sections, Los Alamos Scientific Laboratory, N. Mex., Rept. LA-2122 (1957).
6. W. D. Allen and A. T. G. Ferguson, "The Fission Cross Sections of ^{233}U , ^{235}U , ^{238}U and ^{239}Pu for Neutrons in the Energy Range 0.030 MeV to 3.0 MeV, " Proc. Phys. Soc. (London) **70A**, 573 (1957).
7. G. A. Dorofeev and Yu. P. Dobrynin, "Effective Cross Sections for Fission of U^{233} , U^{235} , Pu^{239} , and Pu^{240} by Neutrons with Energies from 30 keV to 5 MeV, " Atomnaya Energiya (Eng. Trans.) **2**, 9 (1957); Atomnaya Energiya **2**, 10 (1957); J. Nucl. Energy **5**, 217 (1957).
8. S. P. Kalinin and V. M. Pankratov, "Neutron-Induced Fission Cross Sections for Th^{232} and U^{238} in the Energy Range of 3 to 11 MeV and U^{233} , U^{235} , Np^{237} , and Pu^{239} in the Energy Range of 3 to 8 MeV, " Proc. 2nd U. N. Int. Conf. on P. U. A. E. **16**, 136 (1958).
9. G. V. Gorlov, B. M. Gokhberg, V. M. Morozov, G. A. Otroschendo, and V. A. Shigin, "The Fission Cross Sections of ^{233}U and ^{235}U for Neutrons Having Energies Between 3 and 800 keV, " J. Nucl. Energy **12**, 79 (1960).
10. B. Adams, R. Batchelor, and T. S. Green, "The Energy Dependence of the Fission Cross-Sections of ^{238}U , ^{235}U , and ^{239}Pu for Neutrons in the Energy Range 12.6 to 20 MeV, " J. Nucl. Energy **14**, 85 (1961).
11. G. N. Smirenkin, V. G. Nesterov, and I. I. Bondarenko, " U^{233} , U^{235} , and Pu^{239} Fission Cross Sections for 0.3-2.5 MeV Neutrons, " Atomnaya Energiya **13**, 366 (1962); Soviet J. At. Energy **13**, 974 (1962).
12. V. M. Pankratov, "Fission Cross Sections of Th^{232} , U^{233} , U^{235} , Np^{237} , U^{238} , for 5-37 MeV Neutrons, " Soviet J. At. Energy **14**, 167 (1963).
13. J. L. Perkin, P. H. White, P. Fieldhouse, E. J. Axton, P. Cross, and J. C. Robertson, "The Fission Cross Sections of ^{233}U , ^{234}U , ^{235}U , ^{236}U , ^{237}Np , ^{239}Pu , ^{240}Pu and ^{241}Pu for 24 keV Neutrons, " J. Nucl. Energy **19**, 423 (1965).
14. P. H. White, J. G. Hodgkinson, and G. J. Wall, "Measurement of Fission Cross-Sections for Neutrons of Energies in the Range 40-500 keV, " Proc. Symp. Physics and Chemistry of Fission, Salzburg, March 22-26, 1965 (IAEA, Vienna) **1**, 219 (1965).

15. G. D. James, "Fission Cross Section Measurements of ^{233}U , ^{235}U , ^{239}Pu , and ^{241}Pu in the Energy Range from 1 to 24 keV, " Proc. Intern. Conf. Fast Critical Experiments and Their Analysis, Argonne National Laboratory, Oct. 10-13, 1966, Argonne National Laboratory, Argonne, Ill., Rept. ANL-7320 (1966).
16. P. H. White and G. P. Warner, "The Fission Cross Sections of ^{233}U , ^{234}U , ^{236}U , ^{238}U , ^{237}Np , ^{239}Pu , ^{240}Pu , and ^{241}Pu Relative to That of ^{235}U for Neutrons in the Energy Range 1-14 MeV, " J. Nucl. Energy **21**, 671 (1967).
17. J. A. Grundl, "A Study of Fission-Neutron Spectra with High-Energy Activation Detectors - Part I. Detector Development and Excitation Measurements, " Nucl. Sci. Eng. **30**, 39 (1967).
18. V. G. Nesterov and G. N. Smirenkin, "Cross Section Ratios for Fission of U^{233} , U^{235} , and Pu^{239} by Fast Neutrons, " Atomnaia Energiia (Eng. Trans.) **24**, 224 (1968); Atomnaya Energiya **24**, 185 (1968).
19. W. E. Stein, R. K. Smith, and H. L. Smith, "Relative Fission Cross Sections of ^{236}U , ^{238}U , ^{237}Np , and ^{235}U , " Phys. Soc. Conf. Neutron Cross Section Techn., Washington, D. C., National Technical Information Service, Oak Ridge, Tenn., Rept. CONF-680307 (1968), p. 627.
20. W. P. Poenitz, "Measurement of the Ratios of Capture and Fission Neutron Cross Sections of ^{235}U , ^{238}U , and ^{239}Pu at 130 to 1400 keV, " Nucl. Sci. Eng. **40**, 383 (1970).
21. E. Pflutschinger and F. Käppeler, "A Measurement of the Fission Cross Sections of ^{239}Pu and ^{233}U relative to ^{235}U , " Nucl. Sci. Eng. **40**, 375 (1970).
22. M. V. Sabin, Yu. A. Khokhlov, Yu. S. Zamyatin, and I. N. Paramonova, "Ratios of Fast Fission Cross Sections of U^{235} , P^{239} , and Pu^{240} , " Atomnaia Energiia (Eng. Trans.) **29**, 938 (1970); Atomnaya Energiya **29**, 218 (1970).
23. W. K. Lehto, "Fission Cross-Section Ratio Measurements of ^{239}Pu and ^{233}U to ^{235}U from 0.24 to 24 keV, " Nucl. Sci. Eng. **39**, 361 (1970).
24. M. Soleillac, J. Frehaut, J. Gauriau, and G. Mosinski, "Mean Number of Prompt Neutrons and Relative Cross-Sections for the Fission of ^{235}U and ^{239}Pu Between 0.3 and 1.4 MeV, " Proc. 2nd Intern. Conf. Nuclear Data for Reactors, Helsinki, June 15-19, 1970 (IAEA, Vienna) **2**, 145 (1970).
25. W. P. Poenitz and R. J. Armani, "Measurements of the Fission Cross Section Ratio of ^{238}U to ^{235}U from 2-3 MeV, " J. Nucl. Energy **26**, 483 (1972).
26. W. P. Poenitz, "Additional Measurements of the Ratio of the Fission Cross Sections of Plutonium-239 and Uranium-235, " Nucl. Sci. Eng. **47**, 228 (1972).
27. J. W. Meadows, "The Ratio of the Uranium-238 to Uranium-235 Fission Cross Sections from 1 to 5 MeV, " Nucl. Sci. Eng. **49**, 310 (1972).
28. J. W. Meadows, "The Ratio of the Uranium-233 to Uranium-235 Fission Cross Section, " Nuc. Sci. Eng. **54**, 317 (1974).
29. J. W. Behrens, A Painting Process for Making Fission Foils, Lawrence Livermore Laboratory Rept. UCID-15538 (1969).

30. J. W. Behrens, Preparation of Fission Foils for Fission Ionization Chambers Using a Painting Technique, Lawrence Livermore Laboratory Rept. UCRL-51476 (1973).
31. C. J. Rush, "A New Technique for Designing Fast Rise Transistor Pulse Amplifiers," Rev. Sci. Instr. 35, 149 (1964).
32. G. W. Carlson, The Effect of Fragment Anisotropy on Fission Chamber Efficiency, Lawrence Livermore Laboratory Rept. UCRL-75309 Preprint (1974); submitted to Nuclear Instruments and Methods.
33. J. E. Simmons and R. L. Henkel, "Angular Distribution of Fragments in Fission Induced by MeV Neutrons," Phys. Rev. 120, 198 (1960).

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