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ABSOLUTE MEASUREMENTS OF THE ^{233}U (n,f)
CROSS SECTION BETWEEN 0.13 AND 8.0 MeV

by

W. P. Poenitz

Argonne National Laboratory

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Applied Physics Division
Argonne National Laboratory
9700 South Cass Avenue
Argonne, Illinois 60439
USA

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ABSTRACT

The fast neutron fission cross section of ^{233}U was measured absolutely between 0.13 and 8.0 MeV. The absolute cross section values were obtained by low-geometry alpha counting and isotopic dilution analysis of various ^{233}U samples, 2π -detection of the fission fragments with an ionization chamber, and the measurement of the neutron flux with several Black Neutron Detectors. Absolute cross sections were obtained with a 2-3% uncertainty over the most important energy range.

I. INTRODUCTION

The traditional approach of obtaining fission cross section values for isotopes other than ^{235}U was to measure the ratios relative to ^{235}U and to utilize evaluated cross section values for the latter. However, this approach might be of a disadvantage in obtaining cross sections of nuclei like ^{233}U and ^{239}Pu which play a major role in the consideration of alternate breeder cycles and thus are required to be known with high accuracy. Error propagation causes these cross sections to be uncertain by 3-4% based upon a typical 2-3% uncertainty of evaluated ratios and a similar 2-3% uncertainty of evaluated ^{235}U (n,f) cross sections. The problem is significant in the case of ^{233}U where recent ratio measurements differ by ~5% (see Ref. 1). These uncertainty levels, compared with typical 1.5-2.5% uncertainty levels obtained in more recent absolute measurements, suggest that the ^{233}U cross section be absolutely measured over the range of interest for fast breeder reactor design studies. The present measurements employ a technique developed in absolute measurements of the ^{235}U fission cross section.^{2,3}

II. SETUP AND PROCEDURE

Figure 1 shows the experimental setup which is a slight modification of the previously reported experimental arrangement.^{2,3} The modification concern mainly the dimensions and the addition of collimators to further reduce corrections. The fissile samples in the ionization chamber were located ~9 cm from the neutron source and positioned perpendicular to the zero-degree direction of the neutron source reaction. At a distance of ~325 cm, a neutron collimator provided a collimated beam for the 4 π -shielded neutron detector. This detector was positioned at zero-degrees about 500 cm from the neutron source. The main collimator channel had a length of 60 cm, was made of lucite, and had a square cross section. Two different collimator openings were used for the present experiments (cross sections of 0.9217 and 1.6155 cm²). Additional collimators, placed between the main collimator and the neutron detector, did not limit the primary neutron beam but reduced the number of neutrons scattered from the main collimators and from air into the neutron detector. The absolute cross sections were derived from the equation

$$\sigma = \frac{C_f}{C_n} \cdot \frac{\epsilon_n}{\epsilon_f} \cdot \frac{F_c}{n_{U-233}} \cdot \frac{d^2}{l^2} \cdot \prod_i c_i,$$

where C_f and C_n are the observed count rates from the fission chamber and the neutron detector, ϵ_f and ϵ_n are the respective efficiencies, F_c is the cross section area of the main neutron collimator exit which is closest to the neutron detector, n_{U-233} is the number of ²³³U nuclei of the fissile samples. d and l are the distances of the fissile samples and the neutron collimator exit opening from the neutron source. $\prod_i c_i$ is the product of all the correction factors which will be discussed below.

Measurements were carried out with monoenergetic neutrons using the time-of-flight technique for background suppression. C_f and C_n were obtained

Fig. 1. Experimental Arrangement Shown with the Liquid-Scintillator, BND

from the integration over the peaks in the corresponding time-of-flight spectra. Prior subtraction of background time-of-flight spectra had little effect on the results ($<1\%$) except for the measurements with the $D(d,n)$ neutron source where background is primarily due to neutron production in the gas-cell material and high energy neutrons tend to leak through the neutron detector shielding and produce a time-dependent background.

The pulse signals from the fission ionization chamber and the neutron detector were tagged and mixed at an early stage (to assure identical signal processing) and recorded with an on-line computer facility.⁴ Random pulses were obtained from a ^{252}Cf source and a small plastic scintillator which detected the fission fragments. These random pulses were time-correlated with the accelerator pulser frequency by a coincidence unit. The random (but time shifted) pulses were then split on a one-for-one basis and added to the fission and neutron detector signals in a first-stage linear sum unit. They appeared as peaks in the time-of-flight spectra and served as a control for proper pulse-processing from the first to the last stage of the electronics and computer-software. The ratio was found to be one within the uncertainty of the integration procedure ($<1\%$).

Measurements were carried out in a large number of sets, varying the different parameters of the experiment (ϵ_n , ϵ_f , $n_{\text{U-233}}$, F_c , d) and the corrections associated with these parameters. The only parameter not changed in the present experiment was λ . The uncertainty of the measured value of λ contributes little to the uncertainty of the final results. To some extent the considerations by Youden⁵ were followed and several parameters were varied from set to set. However, this was not always possible and only one parameter was changed in some sets.

III. FISSILE SAMPLES AND FISSION COUNTER

Table 1 gives a summary of the fissile samples used in the present experiments. Polished plates of stainless steel (0.025 cm thick, 6 cm in diameter) or molybdenum (0.0125 cm thick) were used as sample backings. The uranium was deposited by molecular plating from a solution of uranyl nitrate in alcohol. The diameter of the deposit was 2.54 cm. The absolute number of ^{233}U nuclei on the sample was determined by low-geometry α -counting at various distances from a surface barrier detector. Most of the counts came from the ^{233}U decay and up to 1.5% came from the ^{232}U decay. The latter were eliminated by pulse-height discrimination. The α -pulse height spectra also contain a low energy tail which amounts to as much as 2% for the thickest samples but was less than 1% for the thin samples.

A value of $(1.591 \pm 0.001) \cdot 10^5$ y was used for the half-life of ^{233}U . This is a weighted average of the existing experimental values given in a table by Jaffey et al.⁶ (a value obtained by EURATOM was recently updated⁷ and the new value was taken into account). A half-life of $2.447 \cdot 10^5$ was used for the α -decay of ^{234}U .⁸ Analysis by isotopic dilution technique of the first six samples of Table 1 yielded mass values differing in the average by only 0.5%. The masses given in the table are the averages of both techniques.

In all absolute measurements, pairs of ^{233}U samples were used as indicated in Table 1. Two samples were mounted back-to-back in about the center of an ionization chamber described by Meadows.⁹ The use of two samples in a back-to-back position reduces the uncertainty for the corrections required for the angular distribution of the fission fragments. A satisfactory separation between pulses from the ^{233}U α -decay and the majority of the fission fragments was achieved with a spacing of ~ 0.8 cm between the samples and the collector

TABLE I. Summary of ^{233}U and ^{235}U Samples Used in the Present Experiments

Sample	Deposit Diameter (cm)	Backing Thickness (cm)	Isotopic Composition (%)						Total Number of Major Isotope Nuclei
			^{232}U	^{233}U	^{234}U	^{235}U	^{235}U	^{238}U	
U3/Mo 1	2.54	0.0125 Mo		99.47	0.183	0.071	0.014	0.266	$1.622 \cdot 10^{18}$
U3/Mo 2	2.54	0.0125 Mo		99.47	0.183	0.071	0.014	0.266	$1.666 \cdot 10^{18}$
U3/Fe 3	2.54	0.025 Fe		99.47	0.183	0.071	0.014	0.266	$2.045 \cdot 10^{18}$
U3/Fe 4	2.54	0.025 Fe		99.47	0.183	0.071	0.014	0.266	$22.274 \cdot 10^{18}$
U3/M 1	2.54	0.025 Fe	.47 ppm	99.92	0.018	0.0015	0.0002	0.056 ppm	$1.510 \cdot 10^{18}$
U3/M 2	2.54	0.025 Fe	.47 ppm	99.92	0.018	0.0015	0.0002	0.056 ppm	$1.510 \cdot 10^{18}$
U3/M 120*	2.54	0.025 Fe		99.54	0.184	0.062	0.013	0.203	$0.655 \cdot 10^{18}$
U3/M 140*	2.54	0.025 Fe		99.54	0.184	0.062	0.013	0.203	$0.765 \cdot 10^{18}$

*These samples were previously used in ratio experiments by Meadows.

plates (0.0125 cm thick molybdenum on one side and 0.025 cm thick stainless steel on the other side). The efficiency for counting fission events was approximately 95%, somewhat larger for the thinner and somewhat less for the thicker samples. The differences in the efficiencies are caused by the differences in the total fission fragment absorption. It also varied with different bias settings for the recording of the time-of-flight spectra. Extrapolation to zero pulse-height was carried out with the assumption that the shape of the fission fragment spectrum at low pulse-heights is similar to equally thick samples of other isotopes like ^{235}U or ^{238}U . The 2-4% extrapolation correction contributed $\sim 1\%$ to the uncertainty of the ^{233}U (n,f) measurements. The physical stability of the samples was assured by α -counting before and after the actual cross section experiment. Changes were less than 0.2% with counting statistics of $\sim 0.1\%$.

IV. THE NEUTRON FLUX MEASUREMENT

Neutron detection has been suggested in the past as the major source of cross section uncertainties and discrepancies. However, extensive studies of existing techniques as well as the development of new detectors has changed this situation. A more detailed discussion appears in the Appendix.

Three Black Neutron Detectors (BND) of different size and scattering material were used in the present experiment to measure the absolute neutron flux. The smallest, a plastic scintillator of a cylindrical shape with an ~ 6.4 cm radius and a height of ~ 15 cm with a single low-noise photomultiplier (RCA 8854), was used at low neutron energies (0.13-2.0 MeV). In the intermediate range (0.4-3.0 MeV) a liquid scintillator with five multipliers distributed over the surface of a cylinder with a height of 31 cm and a radius of ~ 9 cm was used. The scintillator had a decalin base and thus a higher hydrogen content than the plastic scintillators. A large plastic scintillator with 16 photomultipliers (58 DVP) was used in the higher energy range (1.2-8.0 MeV). The height and the diameter of this third BND were both ~ 40 cm. Details of the flux measurement uncertainty estimates are given in the Appendix.

V. CORRECTIONS

The experimental data were corrected for a number of factors which affect the measurements. Counting efficiency and correlated corrections are discussed for the appropriate detectors in Section III and in the Appendix.

A. Differences in the Neutron Flux

Several effects cause the neutron flux measured by the neutron detector to be different from that striking the fissile deposits. The major causes are the loss of neutrons due to scattering in air and the angular distribution of the neutron source reaction. Transmission through air was calculated with ENDF/B-IV¹⁰ cross sections for O₂ and N₂ averaged with the resolution of the present experiment. The actual distance between the fissile deposits and the neutron detector was about 5 m and known with better than ± 1 cm. This distance was reduced by ~ 10 cm in order to account for forward-scattering in front of the BND and detection of these neutrons. The correction was in the range of 3-14% with the largest value at 450 keV, a more typical 8-10% at low energies and 3-5% at higher energies. The maximum uncertainty was estimated as 5% of the correction, based upon an assumed 4% uncertainty of the cross sections and a 2% uncertainty of the effective transmission path.

The angular distribution of the neutron source reaction caused less neutrons to strike the fissile samples than in case for an isotopic source. The opening angle of the neutron detector collimator was very small, thus only the effect on the fissile samples required correction. This correction was obtained by fitting the evaluated angular distributions¹¹ with a polynomial and integrating over the space angle subtended by the fissile deposits ($\sim 8^\circ$). The correction was less than 2% for the $^7\text{Li}(p,n)$ source reaction but up to 15% for the $\text{D}(d,n)$ source. The uncertainty of this correction was estimated to be 5% based on the finding of Liskien and Paulsen that

the mean deviation of the experimental data from this evaluation was 4.5%. An additional correction and associated uncertainty was required owing to the uncertainty in the positioning of the fissile deposits on the zero-degree-neutron direction. The data were corrected for half of the effect of the maximum possible positioning error (up to 0.7% for the highest energy with the D (d,n) reaction) and the same amount was assumed as the uncertainty of this additional correction. Applying the latter correction permits the retaining of a symmetric uncertainty quotation. Scattering in the fissile sample backings, the fission counter wall and the collector plates further reduces the neutron flux detected by the BND. The correction for the transmission through the structural material of the fission chamber was calculated with ENDF/B-IV¹⁰ cross sections of iron averaged with the resolution of the present experiment. The correction was in the 1.5-3.5% range. Recent findings¹³ indicate that the ENDF/B-IV cross section for Fe might be too low by up to 6% in the 1-3 MeV range. This would imply an error of 4% in the correction in the present experiment, thus an error of <0.1% of the cross section. This is small and well covered with an estimated uncertainty of 10% of the correction.

B. In-Scattered Neutrons

The major correction for events caused by scattered neutrons was for neutrons scattered in the sample backings and other parts of the fission detector. The effective sample thickness for these scattered neutrons was much larger than for primary neutrons. The correction for scattering in the construction material of the ionization chamber was based on Monte Carlo calculations by Meadows.¹⁴ The effect was in the 3-6% range with the largest corrections at low energies. The uncertainty was estimated as 10% of the correction and was based upon the uncertainty of the cross sections involved and the uncertainty of the calculations.

The correction for neutrons which scatter in air and produce events in the fission chamber was small (0.3-1.3%).¹⁵ The uncertainty was estimated to be 20% of the correction.

Neutrons scattered in the neutron source structural material have predominantly the energy of source neutrons emitted at 90°. A higher cross section corresponds to the lower neutron energy over most of the range of the present measurements, thus amplifying the effect. Still, mostly due to source anisotropy, the corrections were generally small (~1%) but up to ~5% at low energies where the source reaction and scattering cross sections are more isotropic. The correction was based on Monte Carlo calculations by Meadows¹⁴ and was estimated to have an uncertainty of ~20% of the correction.

Neutron scatter from air into the neutron detector is negligible due to the use of several collimators. The detection probability for neutrons scattered in the collimator material was estimated assuming a ten-times-larger forward than average scattering probability. All neutrons striking the inner collimator walls and those striking the front of the collimator on an area equal to the collimator cross section area were considered as scattering source. The effect was found to be <0.1% due to the small solid angle subtended by the collimator exit opening, and the neutron detector to the scattered neutrons. Transmission through the collimator walls was corrected for using ENDF/B-IV cross sections.

C. Non-Monoenergetic Source Neutrons

The ${}^7\text{Li}(p,n)$ reaction produces a second neutron group above ~2.4 MeV primary energy and a breakup reaction occurs above 3.7 MeV. The additional fission events caused by these neutrons of different energies cannot be separated at all energies by time-of-flight. The BND separates the second neutron group up to ~3 MeV primary neutron energy and always resolves neutrons

from the breakup reaction. As a result the correction is rather small between 3 MeV and the onset of the breakup reaction but the correction increases to about 10% below 3 MeV and to 12% at 4.7 MeV. The correction is dependent on the intensity ratio of the second neutron group (or breakup neutrons) to the first neutron group¹⁶ and the ratio of the cross sections at the respective energies. The uncertainty for this correction was estimated to 5% of the correction for the 2nd neutron group and ~10% of the correction for the breakup reaction. The D (d,n) reaction is monoenergetic in the range of the present measurement and thus not subject to multi-group corrections.

VI. RESULTS, UNCERTAINTIES AND DISCUSSION

The results from the present measurements are summarized in Table 2 and shown in Fig. 2. This table contains the energy, the energy uncertainty, the resolution (all in MeV), the fission cross section, its total uncertainty (both in barns) and the statistical uncertainty in percent. Systematic uncertainties were considered together with the appropriate quantities in the above text and are summarized in Table 3. The data are compared in Fig. 2 with ENDF/B-IV.¹⁰ The present data are significantly higher between 400 and 900 keV and show a different behavior at the second-chance-fission threshold.

TABLE 2. Results of the Present Measurements on ^{233}U (n,f)

E , MeV	ΔE^* MeV	RES^{**} MeV	σ , b	$\Delta\sigma$, b	Statistics, %
.137	.003	.013	2.165	.058	1.5
.152	.003	.016	2.123	.049	1.1
.178	.003	.013	2.142	.051	1.5
.198	.004	.020	2.082	.048	1.4
.220	.003	.023	2.223	.051	1.5
.249	.003	.019	2.172	.046	1.1
.271	.003	.022	2.177	.054	1.8
.298	.004	.022	2.239	.045	1.5
.397	.004	.027	2.071	.042	1.0
.449	.003	.027	1.952	.044	1.2
.497	.004	.025	1.964	.037	0.7
.547	.005	.020	1.931	.039	0.8
.598	.005	.020	1.905	.038	0.8
.702	.005	.021	1.894	.040	1.7
.800	.005	.022	1.921	.036	0.9
.894	.005	.030	1.886	.034	0.4
1.005	.005	.030	1.881	.038	0.8
1.258	.005	.029	1.845	.039	1.1
1.509	.005	.023	1.908	.036	0.6
1.793	.005	.057	1.897	.038	0.7
2.012	.005	.023	1.947	.047	1.5
2.258	.005	.042	1.927	.042	1.0
2.509	.005	.045	1.917	.040	0.8
2.758	.005	.040	1.874	.045	1.5
3.005	.005	.027	1.805	.036	0.7
3.502	.005	.025	1.653	.035	0.8
3.998	.005	.030	1.537	.041	1.7
4.247	.005	.030	1.559	.036	0.9
4.502	.010	.079	1.548	.038	0.8
4.744	.005	.007	1.536	.041	1.3
5.021	.020	.090	1.517	.038	1.0
5.131	.020	.064	1.467	.035	1.0
5.330	.020	.062	1.467	.038	0.9
5.482	.020	.035	1.570	.041	1.1
5.673	.020	.050	1.532	.042	1.2
5.946	.020	.032	1.620	.045	1.3
6.053	.020	.031	1.671	.046	1.0
6.141	.020	.030	1.724	.048	1.1
6.233	.020	.030	1.818	.050	1.2
6.845		.026	2.062	.068	1.5
7.489		.022	2.059	.062	1.6
8.045		.020	2.185	.065	1.8

*Energy scale uncertainty.

**Energy resolution.

TABLE 3. Uncertainty Ranges

Source	Uncertainty Range, %
Mass of Fissile Samples	0.7
Extrapolation	1.0
Distance between Source and Sample	0.6
Collimator Cross Section	0.5
Collimator-source Distance	0.3
Corrections	
Air Transmission	0.2-0.5
Air Scattering	0.2
Chamber Transmission	0.2
Scattering in Chamber	0.4-0.6
Anisotropy of Source	0.2-1.6
Collimator Transmission	0.1-0.4
Scattering in Source	0.2
2nd Neutron Group	0.0-0.5
Chamber Positioning on 0°	0.5-1.4
Fission Fragment Absorption	0.3
Effective Sample Thickness	0.1
Neutron Detection	0.6-1.5

APPENDIX

The Neutron Flux Measurement with the Black Neutron Detector

Neutron detection has been blamed in the past as the major source of cross section uncertainties and discrepancies. However, extensive studies of existing techniques as well as the development of new detectors has changed this situation. Actual proof is difficult in most cases. The problem is that specific measurement techniques are often only applicable to specific experiments and do not permit intercomparison. Thus, intercomparison of direct absolute flux measurements is only possible for a restricted number of techniques or has to be made using the final result e.g. the measured cross section or a monitor calibration. The observed differences in an indirect comparison should be logically explained by:

1. Differences in the one or the other flux measurement technique (and it cannot possibly be said in which).
2. Differences introduced in the different experimental arrangements required by the one or the other flux measurement technique.

Uncertainties from sources other than the flux measurement contribute substantially to the final result (cross section or monitor calibration), thus a comparison of these quantities usually gives little information except the possible upper limit for the uncertainty of the flux measurement.

The Black Neutron Detector^{17,18} was used for absolute flux measurements in the present experiments. Even very conservative estimates of the uncertainty of a measurement with a BND in some energy ranges results in values of <1%. The only other known technique for absolute flux measurements with this level of accuracy is the associated particle technique with the $T(d,n)^4He$ reaction. However, the neutron energy from this reaction (14 MeV) is already in a range for which the BND is not best suited and has an uncertainty of >1%. Still it is interesting to note that a comparison made between both

techniques by G. Grenier¹⁹ yielded agreement within 1%. Grenier also applied the associated particle technique to the $D(d,n)^3\text{He}$ reaction and found agreement with the BND within uncertainty limits of <2%. An extensive study of the flux measurement with the associated particle technique and the BND using the $^3\text{H}(p,n)^3\text{He}$ reaction was carried out at the National Bureau of Standards (Lamaze²⁰ and Meier²¹). The average difference at three energies was 1.2%, well within the uncertainty of the comparison. In this case the accuracy of the associated particle technique is the limiting factor.

Because a direct flux comparison at a <1% level is not available, the estimated uncertainties for BND measurements are considered in detail. Figure 3 shows a pulse-height spectra obtained for 400 keV neutrons. The figure shows that only a small number of pulses are lost due to the detection threshold and thus demonstrates a major advantage of the BND. The pulse height distribution agrees in principle but not in detail with the calculation of light production in the scintillator. The difference has been successfully explained with Poisson-statistics of the photoelectrons produced by the light at the photomultiplied-cathode.²⁰ Little effect can be expected for the efficiency for a spectra as shown in Fig. 3. However, some difference of the efficiency could be expected at lower neutron energies where the "valley" at low pulse-heights is not as pronounced. Figure 4 shows the importance of the only additional parameters required for a Poisson-smear of the originally produced light for 250-keV neutrons. The effective average neutron energy required to produce one photoelectron (PE) must be selected by comparison of the calculated pulse-height spectrum with the experimental spectrum. This is done in Fig. 5. The parameters resulting in the two curves can be considered the limiting values. For 9-keV/PE the efficiency is 98.3% and for 19-keV/PE it is 97.6%. The experimental peak-to-valley ratio agrees better with 19-keV/PE, but the resolution agrees better with 9-keV/PE. The average efficiency

Fig. 3. Pulse Height Spectrum of the Black Neutron Detector for 400 keV Neutrons

Fig. 4. Calculated Pulse Height Spectrum of a BND for Different Values of the Average Amount of Neutron Energy Required to Produce One Photoelectron on the Photocathode of the Multiplier.

Fig. 5. Comparison of the Experimental Pulse Height Spectrum of the BND with Calculated Spectra

from the two calculations (98.0%) shows that 2% of the neutrons are not detected and warrant explanation. A simple calculation shows that 0.14% are transmitted through the detector without any collision, and 0.66% experience only one collision with carbon and thus do not produce enough light to be detected. These 0.8% should be known with better than 5% accuracy (cross section uncertainty), thus, they do not contribute to the uncertainty of the BND efficiency. The uncertainty is entirely determined by the accuracy with which the remaining 1.2% detection losses can be calculated with the Monte Carlo code and the Poisson-Smear. The deviation of the two limiting efficiencies from the average (of 98.0%) provides this measure (0.4%).

Additional uncertainties are due to experimental procedure (scale zero determination, 0.3%), detector parameter and cross section uncertainties (0.1%), and Monte Carlo statistics (0.2%). Thus, the overall uncertainty for the BND detection efficiency at 250 keV is 0.55%. This value, of course, applies only to the specific detector considered here, for which a sufficient "valley" at low pulse-height was achieved. If the spectra is more smeared out and therefore the detection threshold lies in a range of considerable slope in the spectrum, an increased uncertainty must be expected. Also, a larger neutron escape probability, due to a less favorable detector geometry, will cause a lower efficiency and thus a higher uncertainty. Table 4 summarizes the uncertainties applicable at 250 keV.

TABLE 4. Black Neutron Detector Uncertainty at 250 keV

Source	Uncertainty, %
Cross Section Uncertainty*	0.1
Stat. of Monte Carlo Calc.	0.2
Parameter Uncertainty	0.2
Cut-off Energy	0.3
Choice of Poisson Distribution	0.3
Total Uncertainty	0.5

*This effect is small due to the fact that the BND efficiency does not depend upon cross section values in first order. Second order effects in which the cross section uncertainties enter are

Effect	Size	Uncertainty**
Total transmission (no interaction within the BND)	0.14%	0.03%
Loss through entrance channel due to backscatter from carbon (one collision only) or other parts of the detector surface after one collision in carbon	0.66%	0.07%

**Based upon the assumption that the hydrogen and carbon total and differential scattering cross sections are uncertain by 5%, and the atomic density of the scintillator material is uncertain by 15%. Both values were added linearly and are extremely conservative estimates of the uncertainty of these quantities.

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