Precision Measurement of the Total Neutron Cross Section of U^{233} between 0.000818 and 0.0818 ev*

G. J. Safford, W. W. Havens, Jr., and B. M. Rustad
Columbia University, New York, New York and Brookhaven National Laboratory, Upton, New York

(Received November 20, 1959)

The absolute value of the total neutron cross section of U^{233} was measured at neutron energies between 0.000818 ev and 0.0818 ev for two types of samples, a metallic foil and D_2O solutions of uranium nitrate. Balanced solutions of $U^{233}O_2(NO_3)_2$ and $U^{238}O_2(NO_3)_2$ were used to determine the difference between the total cross sections of U^{233} and U^{238}. This value when combined with the relatively small known value of the total cross section for U^{238} gives $\sigma_T(U^{233})=587\pm5$ barns at 0.0253 ev. The measurements on the metallic U^{233} foil agreed with the measured total cross section determined from the liquid solution data to better than 1%, yielding $\sigma_T(U^{233})=586\pm2$ barns at 0.0253 ev.

INTRODUCTION

THE total neutron cross section of U^{233} is one of the basic parameters[1] whose absolute magnitude and shape must be precisely determined to completely characterize the fission process for this element. The total cross section after subtracting the relatively small neutron elastic scattering cross section yields a value of the absorption cross section. This parameter must be known to obtain a value of η (the number of neutrons emitted per neutron absorbed in the fissile material) from pile oscillator measurements, and to obtain a value of the fission cross section from a value of $(1+\alpha)$, (the ratio of the absorption to fission cross section).

A current problem in fission theory has been the apparent need of an extremely large and improbable negative energy resonance to account for the large background cross section in the 0–1.5-ev region for the fissile elements U^{233}, U^{235}, and Pu^{239}.[2] However, recent multilevel fits to the available cross-section data have shown the size of this negative energy resonance can be reduced by attributing part of the large background cross section to interference between resonances of the same spin state.[3,4] Accurate total cross-section data in the 0- to 1.5-ev region for U^{233} are of aid in resolving the relative contributions from a negative energy level and from interference effects between resonant levels on the cross section of this element.

Precise cross-section data for U^{233} is also needed for reactor theory and design. Due to its relatively high fission to radiative capture probability, U^{233} is the most promising breeding fuel for thermal and epithermal reactors which use thorium as a feed material.

Recent published values, as summarized in two sources,[5,6] show that the absorption cross sections obtained from the measurements of the total cross section vary from 577 to 590 barns and have rather large individual errors lying between 2 and 4%. The measured variation[7] of $E^{\frac{1}{2}}\sigma_a$ for U^{233} in the energy region between 0 and 0.1 ev have sufficiently large scatter of data so as not to preclude a significant departure from a $1/v$ behavior in the shape.

The principle difficulties encountered in measurements of the U^{233} total cross section in the thermal and subthermal regions are:

(a) The preparation of a thin U^{233} sample of known thickness and uniformity.

(b) The accurate determination of the isotopic and chemical impurities in the sample.

(c) When a crystal spectrometer is used to determine the cross section in this energy range, corrections must be made for the contamination of first-order reflection with neutrons reflected in higher order. These corrections are so uncertain that until recently, accurate neutron cross-section determinations could not be made with a crystal spectrometer at thermal energy (0.0253 ev).

In the present experiment, the total cross section of U^{233} was measured for neutron energies between 0.000818 ev and 0.0818 ev. Two samples, a U^{233} metallic foil and a D_2O solution of uranyl nitrate, were used to show that the measured cross section was independent of the type of sample used. Improved techniques were used in the preparation of the samples. The higher order contamination of the neutron beam from the crystal spectrometer was reduced to a negligible amount by the use of quartz filters and a mechanical monochrometer.

* This work partially supported by the U. S. Atomic Energy Commission.

[1] For a discussion of the parameters that must be measured to characterize the fission process see, for example, G. J. Safford and W. W. Havens, Jr., Nucleonics 17, No. 11, 134 (1959).

[2] W. W. Havens, Jr., and E. Melkonian, *Proceedings of the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, 1958* (United Nations, Geneva, 1958), Vol. 15, p. 99.

[3] Erich Vogt, Phys. Rev. 112, 203 (1958).

[4] Erich Vogt, Bull. Am. Phys. Soc. 4, 271 (1959).

[5] *Neutron Cross Sections*, compiled by D. J. Hughes and R. B. Schwartz, Brookhaven National Laboratory Report BNL-325 (Superintendent of Documents, U. S. Government Printing Office, Washington, D. C., 1958), 2nd ed., p. 28.

[6] T. S. Green, V. G. Small, and D. E. Glanville, J. Nuclear Energy 4, 409 (1957).

[7] D. J. Hughes and R. B. Schwartz, see work cited in footnote 5, p. 322.

SAMPLE PREPARATIONS

Metallic Foil

A metallic foil, 0.002 in. thick, was rolled from a billet of highly enriched, pure U^{233}, electropolished prior to use, and sealed in aluminum so as to ensure that no oxidation occurred during the measurement.

Upon completion of the transmission measurements, the mean thickness (atoms/cm^2) of the sample was determined from area and weight measurements. A rectangular strip, 0.5000 ± 0.0002 in. by 2.2550 ± 0.0002 in. corresponding to the area upon which the neutron beam was incident, was cut from the foil by a specially constructed precision die. The weight of this strip was determined to be 0.6659 ± 0.00014 g. The mean thickness was corrected for the variations in sample thickness over the above area to obtain the effective sample thickness, used in the calculation of the cross section. The thickness of the foil was measured at 40 points uniformly distributed over the above area to an absolute accuracy of ±0.00001 in. with a supermicrometer. Due to the small amplitude and high frequency of this variation, the correction was found to be small amounting to a 0.05% increase from the measured mean thickness. The final value for the effective foil thickness is $2.3659\times10^{20}\pm0.14\%$ atoms/cm^2.

The isotopic composition of the foil material is given in Table I. Chemical analysis of the foil material showed the high cross-section impurities including the rare earths to be very small. A correction of 0.08% was made on the cross section for the measured chemical impurities. The error in this correction is negligible.

Liquid Samples

Two liquid samples were prepared, one containing, U^{238} nitrate and the other, U^{233} nitrate. The uranium and nitrate concentrations of both solutions were made identical. As the U^{238} cross section is both known and small compared to that of U^{233}, the transmission of the sample containing the U^{238} was used to determine the effective transmission of the nonfissile elements in the U^{233} cell.

The $U^{233}O_2(NO_3)_2$ solution was prepared by dissolving 4.5396 ± 0.0001 grams of oxide free U^{233} metal in DNO_3 in a carefully calibrated standard flask having a volume of 25.00 ± 0.02 cc. The solution was repeatedly heated and then diluted with D_2O to drive off the unreacted DNO_3. Finally, enough D_2O was added to bring the solution to the standard volume. A precision quantitative analysis of the final solution gave 0.1817 ± 0.0004 gram of U^{233} per cc of solution, which is in good agreement with 0.1816 ± 0.0004 gram/cc obtained by the above weight and volume technique. A total nitrate analysis showed the final solution to have a $1.80\pm0.75\%$ normal NO_3 ion concentration.

The $U^{238}O_2(NO_3)_2$ solution was prepared in an identical manner to the U^{233} solution except that 5.46915 ± 0.00045 grams of $U_3^{238}O_8$ were dissolved in the standard volume of 25 ± 0.02 cc. X-ray diffraction analysis and gravimetric determination showed the weighed material to be 99.77% U_3O_8 with regard to possible UO_2 and UO_3 content.[8] This gives rise to a small correction, 0.02% in the sample thickness. The total nitrate analysis of the U^{238} solution was $1.790\pm0.75\%$ normal NO_3 concentration, which matches the total nitrate concentration in the U^{233} solution.

For the neutron transmission measurements, the nitrate solutions were placed in the two matched, boron-free quartz cells whose measured effective thicknesses were 10.00 ± 0.01 mm, where the error quoted is the maximum variation in thickness over the area of the cell. As a check, the cells were filled with D_2O and their transmissions compared at five energies between 0 and 0.1 ev. These measurements confirmed the fact that the cells were matched to 0.1% in thickness. The uranium isotopic purity of the solutions are given in Table I.

The effective thicknesses of the uranium contents of the two cells were, $N(U^{238}\text{ cell})=4.69395\times10^{20}\pm0.22\%$ atoms/cm^2 and $N(U^{233}\text{ cell})=4.69579\times10^{20}\pm0.20\%$ atoms/cm^2 and show the degree to which the cells were matched in uranium content.

The solutions were chemically analyzed for impurities. The largest uncertainty in the U^{233} total cross section resulted from a small HNO_3 contamination in the DNO_3 used in the preparation of the U^{233} sample. An isotopic analysis showed the maximum uncertainty in the final cross section from the source to be $\pm0.8\%$. The final error on the cross section has been broadened to include this uncertainty. The remaining chemical impurities contribute a correction of only 0.1%.

TABLE I. Isotopic purity of samples.

Sample	Isotope	Percent by weight
U^{233} (metal foil)	U^{233}	$99.76\pm0.01\%$
	U^{234}	$0.022\pm0.001\%$
	U^{235}	$0.007\pm0.007\%$
	U^{236}	1 part per million
	U^{238}	$0.21\pm0.01\%$
U^{233} (nitrate solution)	U^{233}	99.35%[a]
	U^{235}	0.07%
	U^{238}	0.55%
U^{238} (nitrate solution)	U^{234}	0.00017%[a]
	U^{235}	0.015%
	U^{238}	99.985%

[a] Mass spectrometer measurement at Oak Ridge National Laboratory. (Numbers quoted have a 95% confidence limit.)

[8] C. J. Rodden, *The Analytical Chemistry of the Manhattan Project* (McGraw-Hill Book Company, Inc., New York, 1950), National Nuclear Energy Series, Plutonium Project Record, Vol. 1, Div. VII, pp. 18, 53–54.

EXPERIMENTAL PROCEDURE AND APPARATUS

The Columbia University neutron crystal spectrometer at Brookhaven National Laboratory Reactor was used for these measurements. Similar types of high precision spectrometers have been described previously in the literature.[9] In the energy range from 0.0253 ev to 0.0818 ev, a germanium single crystal oriented to the (111) plane, for which second-order reflection is forbidden, is used for the neutron monochromator together with a neutron filter composed of large quartz crystal.[10] The measured fractional higher order contamination was less than 0.02% in this energy range. From 0 to 0.0253 ev, a mechanical neutron velocity selector was used to select only the neutrons from first-order reflections.[11] At energies above 0.006 ev, a germanium crystal was used as a spectrometer monochromator. At lower energies, a mica crystal having a larger lattice spacing was used. The total higher-order contamination in the beam after the velocity selector has been shown to be less than 0.5%.[12]

The data for the transmission measurements were taken at each energy in the sequence, (beam open)-(sample in)-(sample in)-(beam open), to minimize the effect of intensity drifts in the neutron beam from the reactor. More than 300 such cycles were taken at each energy to obtain the desired statistical accuracy of about 0.1%. The transmission data at each energy consisted of 4 or 5 individual measurements spaced as much as 2 weeks apart. In all cases, the resulting cross sections were found to agree to within statistical accuracy, which was less than 0.5% for most of the individual measurements. This procedure showed that the data were reproducible.

The area of the neutron beam incident upon the samples was determined by careful collimation of the beam with cadmium and lithium fluoride slits. The center of each sample was aligned relative to the neutron beam to an accuracy of 0.001 in.

Other possible errors arising from spectrometer operation such as energy determination, resolution, background, and dead time have been investigated and were found negligible.

RESULTS

The cross section for U^{233} in the liquid cells was calculated from the measured transmissions of the samples containing the $U^{238}O_2(NO_3)_2$ and $U^{233}O_2(NO_3)_2$, the measured thicknesses of the quartz cells, and the measured number of uranium atoms per cc of solution, which was the same for the two cells. The ratio of the measured transmissions of the two cells gives $T(U^{233})/T(U^{238})=\exp\{-n[\sigma_T(U^{233})-\sigma_T(U^{238})]\}$, where n is the number of atoms/cm^2 of uranium in each cell. A value of $\sigma_T(U^{238})$ was added to the resulting $[\sigma_T(U^{233})-\sigma_T(U^{238})]$ to determine $\sigma_T(U^{233})$. The values for the U^{238} total cross section were obtained by adding a $1/v$ capture term, which was normalized to a 2.74 ± 0.02 barns at 0.0253 ev, to an energy-independent, free-atom scattering cross section of 9.5 ± 0.5 barns.[13] This assumption is valid in view of the absence of nearby resonances in U^{238}.

The Remington Rand 409.2R computer at Brookhaven National Laboratory was used to calculate the sample transmissions for each data collection cycle and the statistical variation in the data based on the counting statistics. Those runs with a standard deviation greater than 2.5 times the mean were discarded in accordance with the Chauvenet criterion.[14] The number of sets which had a deviation greater than 2.5 times the average was less than one in 200, as is expected statistically. A new calculation of the mean and the standard deviation was then made. All the resulting sets of data were satisfactory on the basis of Pearson's chi-squared test. All errors quoted unless otherwise stated are one standard deviation.

The effects of liquid diffraction and of the interference terms in the coherent scattering cross section of the UO_2^{++} ion were calculated for both samples. Due to the degree to which the U^{233} cross-section cells were matched, the combined correction on the cross section from these effects is less than 0.07%.

The measured values of the total cross sections for the foils and liquid samples are given in Table II. The function, $E^{\frac{1}{2}}\sigma_T$ vs E, for U^{233} are plotted in Fig. 1 for both the liquid and metallic samples. The errors, shown on this graph and listed in Table II, include both statistical errors and errors in the determination of the sample thickness.

The lines shown in Fig. 1 are weighted least-squares fits to a straight line for the foil data between 0.005 ev and 0.06 ev and for the liquid sample data between 0.000818 ev and 0.0818 ev. These fits show that $E^{\frac{1}{2}}\sigma_T(U^{233})$ is constant in this energy range within the statistics of the data.

The values of the total cross section determined with the liquid samples and with the metallic foils agree within the accuracy of the experiment between 0.005 ev and 0.0600 ev. Below 0.005 ev, the results for the metallic foils are systematically smaller than those for the liquid samples. The measured total scattering cross section of U^{233} is constant and has a value of 12.5 ± 0.5 barns in the thermal[15] region, and the coherent scattering, while unknown, could be this large. The Bragg

[9] V. L. Sailor, H. L. Foote, Jr., H. H. Landon, and R. E. Wood, Rev. Sci. Instr. **27**, 26 (1956).
[10] B. M. Rustad, J. T. Wajima, and E. Melkonian, Bull. Am. Phys. Soc. **4**, 245 (1959).
[11] N. Holt, Rev. Sci. Inst. **28**, 11 (1957).
[12] J. A. Moore, J. Rush, and B. M. Rustad, Bull. Am. Phys. Soc. **4**, 245 (1959).
[13] D. J. Hughes and R. B. Schwartz, see works cited in footnotes 5 and 7.
[14] Yardley Beers, *Introduction to the Theory of Error* (Addison-Wesley Publishing Company, Inc., Reading, Massachusetts, 1953), pp. 24–25.
[15] S. Oleksa, Phys. Rev. **109**, 1645 (1958).

TABLE II. The measured U^{233} total neutron cross sections.

Neutron energy E (ev)	Liquid sample $\sigma_T(U^{233})-\sigma_T(U^{238})$ (barns)	$\sigma_T(U^{238})$ (barns)	$\sigma_T(U^{233})$ (barns)	$E^{\frac{1}{2}}\sigma_T(U^{233})$ (barns$-$ev$^{\frac{1}{2}}$)	Metal foil $\sigma_T(U^{233})$ (barns)	$E^{\frac{1}{2}}\sigma_T(U^{233})$ (barns$-$ev$^{\frac{1}{2}}$)
0.000818	3225.6±28.8	24.74±0.51	3250.3±28.8	92.96±0.83	3194.6±8.7	91.37±0.25
0.00128	2612.1±23.4	21.68±0.51	2633.8±23.4	94.23±0.84	2552.4±6.1	91.42±0.22
0.00291	1694.6±16.4	17.58±0.50	1712.1±16.4	92.36±0.89	1694.9±3.5	91.43±0.19
0.00511	1277.3±11.4	15.60±0.50	1292.9±11.4	92.43±0.82	1297.0±2.8	92.72±0.20
0.00909	952.3± 8.4	14.07±0.50	966.4± 8.4	92.14±0.80		
0.0253	571.7± 4.9	12.24±0.50	584.0± 5.0	92.90±0.80	584.8±1.6	93.03±0.26
0.0300	526.7± 4.6	12.02±0.50	538.8± 4.6	93.33±0.81	537.3±1.6	93.07±0.27
0.0350	491.5± 4.3	11.83±0.50	503.3± 4.3	94.17±0.81	499.7±1.6	93.49±0.31
0.0375	477.1± 4.3	11.75±0.50	488.9± 4.3	94.78±0.85	479.0±1.6	92.86±0.31
0.0425	447.2± 3.9	11.61±0.50	458.8± 3.9	94.60±0.82	451.7±1.7	93.14±0.37
0.0449	438.7± 3.8	11.56±0.50	450.3± 3.9	95.42±0.83	440.0±1.8	93.32±0.39
0.0500	409.4± 3.7	11.45±0.50	420.9± 3.7	94.12±0.84	423.3±2.1	94.66±0.49
0.0550	387.0± 3.7	11.36±0.50	398.3± 3.7	93.42±0.88		
0.0600	373.7± 3.4	11.28±0.50	385.0± 3.5	94.32±0.86	379.2±1.3	92.90±0.32
0.0650	346.0± 3.4	11.21±0.50	357.2± 3.4	91.08±0.88		
0.0700	343.0± 2.9	11.15±0.50	354.1± 3.0	93.70±0.80		
0.0725	328.6± 2.9	11.12±0.50	339.8± 2.9	91.49±0.80		
0.0750	334.5± 2.9	11.09±0.50	345.6± 2.9	94.70±0.82		
0.0818	309.6± 3.6	11.02±0.50	320.6± 3.6	91.72±1.06		

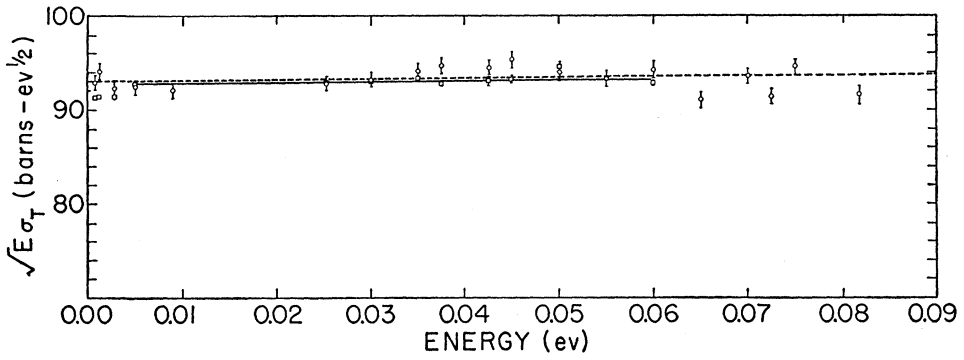

FIG. 1. The variation of $E^{\frac{1}{2}}\sigma_T(U^{233})$ with neutron energy E. ○ ≡Liquid cell data. □ ≡Metal foil data. The dashed line represents the weighted least-squares fit to a straight line for the liquid cell data, and the solid line the weighted least-squares fit to a straight line for the metal foil data.

cutoff for U^{233} is expected to occur near 0.00291 ev, the observed cutoff of U^{238}. Thus below 0.00291 ev, the cross section measured with the foils might be expected to lie lower than that for the liquid samples by as much as 12 barns. However, below 0.00291 ev, the difference in cross section for the liquid and metal samples are not larger than 2.5 times the standard deviation; and, no attempt was, therefore, made to correct the foil measurements for the U^{233} coherent scattering cross section.

The least squares analysis of the liquid sample data yields a final value of 587±5 barns at 0.0253 ev for the U^{233} total cross section. The similar analysis of the metal foil data yields a value of 586±2 barns.

ACKNOWLEDGMENTS

The authors are particularly indebted to Mr. Neil Case of the Oak Ridge National Laboratory, who prepared the liquid U^{233} sample, and to Mr. John Cooper, who analyzed this sample. We are indebted to Dr. J. A. Harvey of Oak Ridge National Laboratory for supplying the U^{233} foils for this measurements.

We also wish to thank Dr. C. J. Rodden of the Atomic Energy Commission New Brunswick Laboratory for his invaluable advice on the preparation of the liquid samples and his analysis of the U_3O_8 samples. We are also indebted to Dr. H. L. Finston and the members of the Hot Laboratory at Brookhaven National Laboratory for performing the x-ray analysis of the U_3O_8 and the total nitrate ion determinations on the liquid samples. We are grateful to Mr. R. J. Graeser and Mr. S. M. Puri for their aid in taking the data, and to Mr. I. Schroder and Mr. G. Mehta for their assistance in performing calculations and analyzing the data.