A DIRECT COMPARISON OF DIFFERENT EXPERIMENTAL TECHNIQUES FOR MEASURING NEUTRON CAPTURE AND FISSION CROSS SECTIONS FOR ^{239}Pu*

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A comparison of the results of two different experimental methods of measuring the neutron absorption and fission cross sections for ^{239}Pu is made. These measurements were normalized at thermal energy and extend to 200 keV. The ratio of the neutron capture to fission cross section for ^{239}Pu derived in these two experiments is shown to be in good agreement.

(^{239}Pu; fission, absorption; measurement; comparison; cross sections)

Introduction

Measurements of the neutron absorption σ_a and neutron fission σ_f cross sections, and thereby the neutron capture σ_c cross section, have been performed at ORNL using two different techniques. These experiments covered the neutron energy region from 0.02 eV to 200 keV and represent a part of the experimental program on ^{239}Pu at ORNL. Two different techniques were used to serve as a guide for further experimentation at ORNL and to provide a measure of confidence in the measurements of $\bar{\sigma}_c/\bar{\sigma}_f$.

Several aspects of the two experiments were essentially the same, such as the measurement of the neutron flux; however, the performance and analysis of these experiments were independent as well as the techniques for observing fission and absorption events. The values $\bar{\sigma}_a$, $\bar{\sigma}_f$, and $\bar{\sigma}_c/\bar{\sigma}_f$ derived in these experiments are given in this paper along with the respective values from ENDF/B-IV MAT-1264.[1] Also included are the values σ_c/σ_f for ^{239}Pu from the evaluation of Sowerby and Konshin.[2]

Similarities in the Two Experiments

Both of the experiments described in this paper were performed at ORNL using the Oak Ridge Electron Linear Accelerator (ORELA) to produce the source of neutrons in bursts from 5 to 30 nsec wide. The energy of the neutrons was measured by the time-of-flight technique. The neutron flux was measured in each experiment using a parallel plate (pulse) ionization chamber filled with BF_3. The prescription for the $^{10}B(n,\alpha)$ cross section used to extract the energy dependence of the neutron flux was that given in ENDF/B-III and was the same as that suggested by Sowerby and Patrick.[3] The energy range 0.02 eV to 200 keV was covered in one run in both experiments; this approach eliminates the problems encountered in internormalizing runs obtained under different experimental conditions and which cover a common but narrow energy interval. Normalization of the present data sets was performed in the thermal energy region using values of σ_f and σ_a from ENDF/B-III for ^{239}Pu.

Description of the Two Experimental Methods

One experiment (method 1) was performed flight path of about 20 m. A 3-in. diameter ^{239}Pu sample having a mass of 0.24 g/cm^2 was used. Fission events were recorded using pulse-shape discrimination to detect fission neutrons and a separate detector "total energy detector" (similar to a Moxon-Rae detector) was used to measure the prompt gamma rays following neutron absorption in the sample. In the other experiment (method 2), the ^{239}Pu (0.03 g/cm^2) was contained in a multi-plate pulse ionization chamber. A large liquid scintillator was used with a 40 m flight path to detect the prompt gamma rays resulting from neutron absorption in the sample, and pulses from this scintillator system in coincidence with pulses from the fission chamber were defined as fissions.

Auxiliary experiments were performed to test various features of the experiments. For example, the neutron (pulse-shape discrimination) detector was run in coincidence with the ^{239}Pu fission chamber in a series of measurements.[4] Also, measurements of the neutron flux were made using ^6Li glass.

Presentation of the Data

The table shows the average values of $\bar{\sigma}_a$, $\bar{\sigma}_f$, and $\bar{\sigma}_c/\bar{\sigma}_f$ derived from the two independent experiments. Also shown are the cross sections from ENDF/B-IV and the values $\bar{\sigma}_c/\bar{\sigma}_f$ for ^{239}Pu taken from the evaluation of Sowerby and Konshin. In comparing the data shown in the table an average difference refers to the sum over a number of energy intervals of the percent difference between the values divided by the number of intervals. In the cases shown in the table where explicit results for $\bar{\sigma}_c$ and $\bar{\sigma}_f$ are not given values of $\bar{\sigma}_c/\bar{\sigma}_f$ for larger intervals than those in the table are obtained using σ_a from ENDF/B-IV along with the appropriate $\bar{\sigma}_c/\bar{\sigma}_f$ to yield $\bar{\sigma}_c$ and $\bar{\sigma}_f$ for each subinterval. These latter values of $\bar{\sigma}_c$ and $\bar{\sigma}_f$ are then averaged over the large interval. The uncertainties shown for $\bar{\sigma}_f$ and $\bar{\sigma}_a$ in the table for method 2 represent the precision of the experiments. For $\bar{\sigma}_c/\bar{\sigma}_f$ the uncertainties shown include the known uncertainties except those due to errors in the cross sections below 0.4 eV used in the normalization. An examination of the table shows that the results $\bar{\sigma}_c/\bar{\sigma}_f$ from the two experiments overlap within their uncertainties, and in fact about 70% of the results for method 1 fall within the uncertainty shown for method 2.

The results of two experiments for $\bar{\sigma}_f$ and $\bar{\sigma}_a$ agree within 0.7% and 0.3%, respectively, for the neutron energy range from 0.1 to 1.0 keV, and the average difference between the two results for $\bar{\sigma}_f$ and $\bar{\sigma}_a$ are 1.9% and 2.9%, respectively. Above 1 keV the neutron cross sections obtained in method 1 are about 4% lower than those derived in method 2. This is thought to be due to difficulties in the measurement of the neutron flux for method 1 at the time of these experiments.

For the 28 intervals shown in the table, the present two experimental values of $\bar{\sigma}_c/\bar{\sigma}_f$ have an average difference of about 6%. A comparison of the average values of $\bar{\sigma}_c/\bar{\sigma}_f$ obtained in the present experiments for the intervals 0.1 to 1, 1 to 10, and 10 to 100 keV shows that they differ by 2.3, 1.5, and 2.3%, respectively, and the results for method 2 are higher in each

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Average Neutron Cross Sections for ^{239}Pu, 0.1 to 200 keV

$$\bar{\sigma} = \frac{\int_{E1}^{E2} \sigma(E)dE}{E2 - E1}$$

E1 - E2 (keV)	Method 2[(a)] $\bar{\sigma}_f$[(c)] barns	Method 2[(a)] $\bar{\sigma}_a$ barns	Method 2[(a)] $\bar{\sigma}_c/\bar{\sigma}_f$[(d)]	Method 1[(b)] $\bar{\sigma}_f$ barns	Method 1[(b)] $\bar{\sigma}_a$ barns	Method 1[(b)] $\bar{\sigma}_c/\bar{\sigma}_f$[(d)]	Sowerby and Konshin $\bar{\sigma}_c/\bar{\sigma}_f$	ENDF/B-IV Mat 1264 $\bar{\sigma}_f$ barns	ENDF/B-IV Mat 1264 $\bar{\sigma}_a$ barns	ENDF/B-IV Mat 1264 $\bar{\sigma}_c/\bar{\sigma}_f$
0.1 - 0.2	17.96 ± .04	33.66	0.87 ± .015	18.41	34.45	.871 ± .052	.845 ± .077	18.20	34.97	.92
0.2 - 0.3	17.90 ± .05	34.69	0.94 ± .010	17.77	34.24	.927 ± .056	.912 ± .094	17.50	34.67	.98
0.3 - 0.4	8.48 ± .03	18.31	1.16 ± .014	8.43	18.12	1.15 ± .069	1.15 ± .099	8.54	18.07	1.12
0.4 - 0.5	9.40 ± .05	13.56	0.44 ± .013	8.47	13.50	.426 ± .026	.483 ± .058	9.67	14.04	.45
0.5 - 0.6	15.46 ± .09	26.54	0.72 ± .040	15.64	26.87	.718 ± .043	.704 ± .069	15.73	27.10	.72
0.6 - 0.7	4.55 ± .03	11.57	1.54 ± .040	4.38	10.90	1.488 ± .089	1.673 ± .133	4.57	11.29	1.47
0.7 - 0.8	5.34 ± .07	10.52	0.97 ± .017	5.54	10.47	.890 ± .053	.973 ± .087	5.55	10.68	.93
0.8 - 0.9	5.10 ± .03	9.30	0.82 ± .025	5.02	8.99	.790 ± .047	.778 ± .101	5.20	9.50	.83
0.9 - 1.0	7.83 ± .14	13.23	0.70 ± .026	8.02	13.43	.675 ± .041	.717 ± .077	8.11	13.81	.70
1 - 2	4.52 ± .02	8.31	0.84 ± .013	4.45	8.02	.802 ± .048	.927 ± .093	4.60	8.37	.82
2 - 3	3.32	6.63	1.00	3.25	6.41	.972 ± .058	1.108 ± .103	3.40	6.76	.99
3 - 4	3.04	5.24	0.72 ± .066	2.95	5.13	.738 ± .043	.895 ± .086	3.08	5.42	.76
4 - 5	2.37 ± .01	4.44	0.87 ± .040	2.35	4.30	.831 ± .050	.821 ± .079	2.41	4.68	.86
5 - 6	2.32 ± .02[(e)]	4.23[(e)]	0.82 ± .046[(e)]	2.13	3.85	.807 ± .048	.867 ± .084	2.27	4.14	.82
6 - 7	2.05 ± .02	3.68	0.79 ± .040	2.01	3.51	.745 ± .045	.816 ± .086	2.06	3.65	.77
7 - 8	2.11 ± .01	3.45	0.64 ± .022	2.03	3.33	.642 ± .038	.629 ± .073	2.09	3.44	.65
8 - 9	2.28 ± .01	3.51	0.54 ± .022	2.06	3.17	.537 ± .032	.575 ± .064	2.17	3.35	.54
9 - 10	1.92 ± .02	2.97	0.55 ± .022	1.76	2.83	.606 ± .036	.617 ± .067	1.95	3.08	.58
10 - 20	1.78 ± .02	2.63	0.48 ± .022			.486 ± .029	.466 ± .05	1.77	2.64	.50
20 - 30	1.64 ± .02	2.22	0.35 ± 0.18			.332 ± .066	.373 ± .04	1.67	2.26	.36
30 - 40	1.61 ± .01	2.09	0.30 ± .041			.247 ± .049	.296 ± .03	1.61	2.07	.28
40 - 50	1.54 ± .02	1.94	0.26 ± .020			.254 ± .051	.242 ± .03	1.59	2.01	.27
50 - 60	1.66 ± .01	2.03	0.23 ± .020			.246 ± .049	.206 ± .03	1.62	2.06	.26
60 - 70	1.62 ± .03	1.99	0.23 ± .025			.220 ± .044	.182 ± .025	1.61	1.99	.23
70 - 80	1.64 ± .04	1.95	0.19 ± .025			.215 ± .043	.165 ± .025	1.63	1.97	.21
80 - 90	1.52 ± .02	1.85	0.22 ± .030			.200 ± .040	.159 ± .03	1.56	1.88	.21
90 - 100	1.54 ± .06	1.80	0.17 ± .045			.138 ± .028	.160 ± .03	1.57	1.82	.16
100 - 200	1.61 ± .02	1.86	0.15 ± .010			.148 ± .030	.170 ± .028	1.53	1.74	.14

(a) Fission chamber plus large liquid scintillator.

(b) Solid sample, "total energy detectors," and fast-neutron detection.

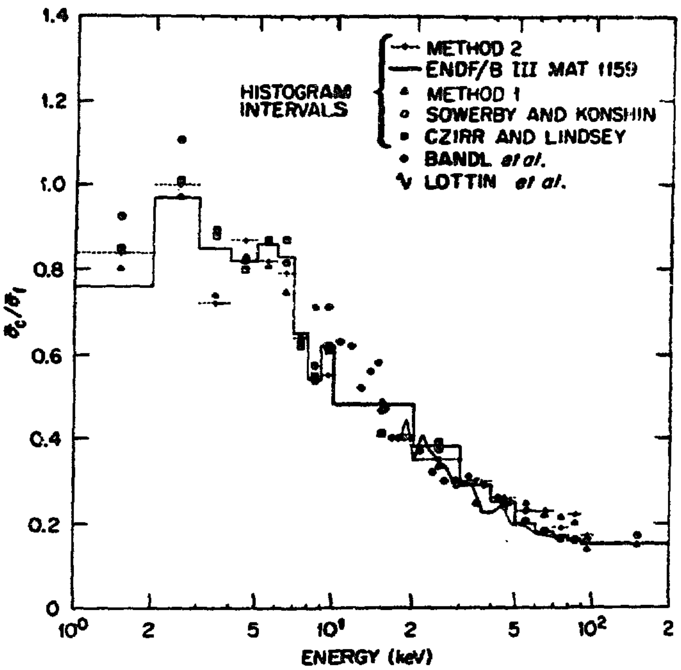
(c) Experimental precision, $S^2 = \frac{1}{N(N-1)} \sum_{i=1}^{N} (x_i - \bar{x})^2$

(d) Includes all known experimental errors.

(e) The aluminum resonance at 5.9 keV perturbs the measurement at this energy.

interval. The ENDF/B-IV values for ^{239}Pu, MAT 1264 were based in part upon the data obtained in the present two experiments.

The relation of the present results for $\bar{\sigma}_c/\bar{\sigma}_f$ for ^{239}Pu and other experimental data can be summarized by comparing them with the results obtained in the evaluation of Sowerby and Konshin. From 0.1 to 40 keV the average difference between the present values (either set for $\bar{\sigma}_c/\bar{\sigma}_f$ of ^{239}Pu and those of Sowerby and Konshin is about 7%. For the energy intervals 1 to 10 keV and 50 to 100 keV, the present average values of $\bar{\sigma}_c/\bar{\sigma}_f$ are 10% lower and 17% higher, respectively, than those obtained by Sowerby and Konshin.



Ratio of $\bar{\sigma}_c/\bar{\sigma}_f$ for ^{239}Pu, 1.0 to 200 keV.

The figure shows a plot of $\bar{\sigma}_c/\bar{\sigma}_f$ for ^{239}Pu obtained in the present two experiments along with those derived by Sowerby and Konshin. Experimental values measured by Czirr and Lindsey,[5] Bandl et al.,[6] and Lottin et al.[7] are also shown on the figure. Values from ENDF/B-III, MAT 1159 are also shown in the figure. Note that the earlier version (III) of ENDF/B shown in the figure follows very closely the results of the evaluation of Sowerby and Konshin above 10 keV and both of these data sets are systematically lower than the results of the present two experiments. These two above evaluations follow the experimental values of Lottin above 50 keV. Earlier measurements of $\bar{\sigma}_c/\bar{\sigma}_f$ by Hopkins and Diven[8] (not shown in the figure) yield a value of .15 for $\bar{\sigma}_c/\bar{\sigma}_f$ for ^{239}Pu at about 60 keV, which supports the two evaluations shown in the figure. The data of Bandl et al. were normalized in the energy region from 40 to 50 keV and thus provide information on the energy dependence of $\bar{\sigma}_c/\bar{\sigma}_f$ only.

Summary

The neutron cross sections $\bar{\sigma}_f$ and $\bar{\sigma}_a$ and especially the ratio $\bar{\sigma}_c/\bar{\sigma}_f$ for ^{239}Pu obtained by these two experiments agree to within a few percent over the energy range of the experiments. In addition, these two results for $\bar{\sigma}_c/\bar{\sigma}_f$ for ^{239}Pu have been shown to be in good accord with the results of other measurements as reflected in the evaluation by Sowerby and Konshin.

Many factors influence the choice of detector systems for the simultaneous measurement of neutron capture and fission cross sections. The authors conclude that for the pursuit of high accuracy measurements of $\bar{\sigma}_c/\bar{\sigma}_f$ on ^{239}Pu a fission chamber used in conjunction with total energy detectors represents a logical choice at ORELA. The total energy detectors are small and can be easily moved from one flight station to another in order to optimize the experimental conditions. Although the detection efficiency of the large liquid scintillator is larger (about a factor of 10 to 20 in the present case) than that of the total energy detector, a gain in the efficiency of the total energy detector system can be made by using additional detectors.

In the present experiments the signal-to-time-dependent background ratio for the large liquid scintillator system is about a factor of 2 to 10 less than that observed for the total energy detector system. Some of the background in the large liquid scintillator can be reduced by the dividing of the tank into optically separated sections and requiring that at least two of these sections detect the event; however, this makes the detector more sensitive to changes in the capture gamma-ray cascade with neutron energy and it also decreases the efficiency of this detector. The design and mode of use of the total energy detector minimizes possible changes in its response as the capture gamma cascade changes.

A fission chamber is preferred for use because the fission detection efficiency can approach unity (~ 95% efficiency). As the efficiency for fission detection approaches unity, all the fission events are identified and capture is measured directly.

The direct measurement of the neutron capture rate simplifies the normalization of $\bar{\sigma}_c$ and eliminates a large part of the uncertainty in $\bar{\sigma}_c/\bar{\sigma}_f$ when this ratio departs from that used in the normalization. In order to achieve the large efficiency (~ 95%) for fission fragment detection, the thickness of the fissile isotope is limited to about 100 μg/cm² and for ^{239}Pu considerations of the alpha particle decay rate limit the total amount of the isotope that can be used with a single detector system. The present fission chamber contained a total of 1.4 g of ^{239}Pu and had an efficiency of about 50%. Investigations of signal-to background ratios with the total energy detector show that measurements of $\bar{\sigma}_c/\bar{\sigma}_f$ for ^{239}Pu using 0.1 g quantities of the isotope are feasible.

References

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[1]Evaluated Nuclear Data File (ENDF/B) of the National Neutron Cross Section Center. A detailed list of the evaluators for the particular isotope (defined with a Mat number, Mat 1159 for ^{239}Pu, ENDF/B, Version III) is given in File 1 of the data tape. The ENDF/B data tape is available from National Neutron Cross Section Center. The average neutron cross sections from the ENDF/B tape used in the present work were obtained using the processing code SUPERTOG, R. Q. Wright et al., "SUPERTOG: A Program to Generate Five-Group Constants and Pu Scattering Matrices from ENDF/B," ORNL-TM-2679 (1969).

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