FISSION CROSS-SECTIONS OF ^{235}U and ^{239}Pu AVERAGED OVER ^{252}Cf NEUTRON SPECTRUM*

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Abstract—A series of measurements have been carried out to derive values for the spectrum-averaged fission cross-section of ^{235}U and ^{239}Pu for ^{252}Cf fission neutrons. Two nearly identical target foils were mounted on either side of a Cf source (10^7 neutron/sec) in a compensated beam geometry. Fission fragments passing through limited solid angle apertures were recorded from each foil by solid-state track-etch techniques. The Cf neutron source strength was calibrated in manganese bath relative to the standard source NBS-II. Values of 1.215 ± 0.022 barn for ^{235}U and 1.790 ± 0.041 barn for ^{239}Pu were obtained for the fission cross-sections, corresponding to a ratio value of 1.473 ± 0.041.

1. INTRODUCTION

Valuable information in nuclear data validation is provided by cross-section measurements over the Cf spontaneous fission neutron spectrum. In comparing the integral results with predictions of data differential in energy, the shape of the fission neutron spectrum is required. For ^{235}U and ^{239}Pu which have slowly varying cross-sections over fission neutron energies, a detailed knowledge of the fission spectrum is not necessary.

Such integral measurements are simple to perform and the results depend primarily on the neutron source strength, fission deposit masses and geometric measurements. The measurements reported here, take advantage of the high accuracy obtainable by manganese bath source calibration and solid-state track-etch techniques. In addition, an integral experiment using limited solid-angle apertures is less sensitive to the uncertainties in fission fragment emission anisotropy than measurement using monoenergetic sources.

2. EXPERIMENTAL METHOD

The experimental apparatus and procedure was, in principle, identical to that described previously for photoneutron sources (Davis *et al.*, 1978) Reaction rates were determined from the number of fission events recorded on polyester film, the geometry of the limited solid angle apertures and the duration of the exposure. The average neutron flux was determined by comparing the ^{252}Cf emission rate relative to NBS-II, the internationally compared Ra–Be photoneutron source. The number of target nuclei was determined from mass assays of the foil deposits.

2.1. *Neutron source calibration*

The Cf source was supplied by the Savannah River Laboratory under its University Loan Program. It was fabricated by uniformly electrodepositing californium hydroxide on 90% Pt–10% Ir cathodes 3.0 cm in length. After they were flamed to form Cf_2O_3 the cathodes were sealed inside Pt–Ir cells with silver braze. The cells were encapsulated in an outer Pt–Ir shell and the entire source sealed inside a stainless steel capsule 4.8 cm long by 0.48 cm in diameter. The positioning and uniformity relative in the capsule surfaces were verified to ± 0.2 mm by a gamma scan with a collimated NaI scintillation detector.

The aspherical shape of the Cf source results in an anisotropic neutron field. Neutrons originally emitted along the length of the source are scattered isotropically, increasing the flux along the equatorial plane while depressing the polar flux.

The shape and magnitude of the polar depression was measured using a long counter set at varying distances from the source. The difference between the polar and equatorial counting rates are plotted versus source distance in Fig. 1. At short distances, the polar depression falls entirely in the shallow of the detector's active surface area. As one moved further away, the count rate difference dropped off predictably as $1/(\text{distance})^2$. Based on these rotation measurements, the ratio for 4π emission to emission in a direction
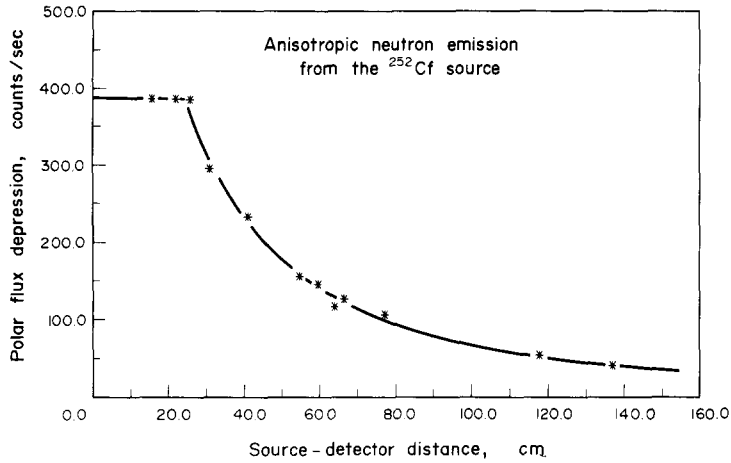
Fig. 1. Anisotropic neutron emission from Cf source.

perpendicular to the cylinder axis (the normal source–target arrangement) was found to be 0.9991 ± 0.0009 (assuming a long counter intrinsic efficiency of 0.1 ± 0.1%).

The Cf source was calibrated in manganese bath against the secondary Ra–Be photoneutron standard, NBS-II. The Cf source was calibrated before and after the fission rate experiments and had a nominal value of $1.0 \times 10^7 \, \text{sec}^{-1}$. The source strength was corrected for decay using a half-life of 2.638 yr. For comparison with differential cross-section data, the Grundl and Eisenhauer (1975) evaluation of the ^{252}Cf spectra was used. As discussed by Gilliam and Knoll (1975), the source strength of NBS-II was taken as $1.174 \times 10^6 \, \text{sec}^{-1}$ ± 0.5%, referred to June 1972.

The Michigan manganese bath facility has been previously described in detail (Gilliam, 1975; Robertson et al., 1975) Small corrections applied to the manganese calibration results include bulk penetration of the bath. A long counter was chosen for this measurement for its flat response over the measured energy range. The ANISN (Engle, 1967) computed bath leakage spectrum for Cf neutrons is shown along with the long counter instrinsic efficiency energy dependence in Fig. 2. Leakage measurements at various distances from a bare source. For a bath radius of 50 cm and a solution density of 1.31 g/cm^3, leakage rates of 0.24 ± 0.03% and 0.0 ± 0.3% were measured for Cf and Ra–Be neutrons, respectively.
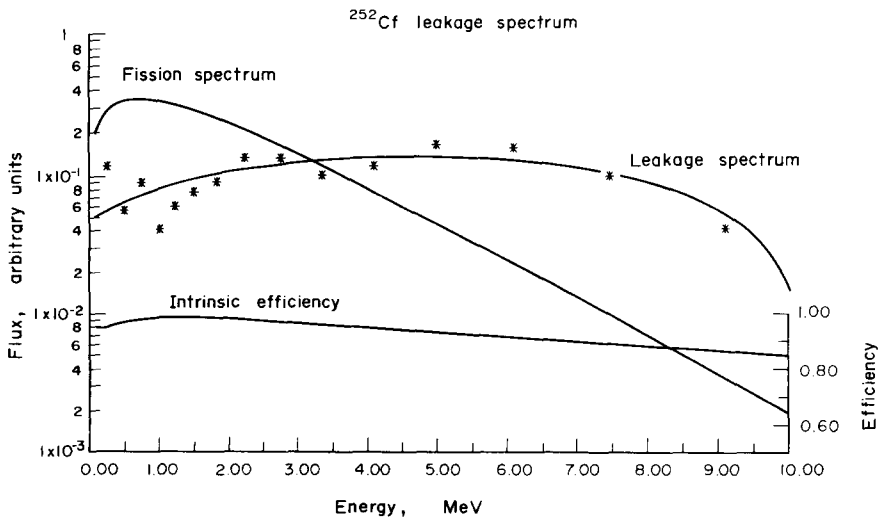

Fig. 2 Manganese bath leakage spectrum from Cf neutrons.

3. DATA ANALYSIS, CORRECTIONS AND RESIDUAL ERRORS

3.1. Fission counting efficiency

The cross-section was derived from the sum of the fluxes at the two deposits and combined fission rates. Similar uncertainties were experienced here in measuring the source–detector positions as were experienced for photoneutron sources.

A correction of (1.38 ± 0.17)%, required to account for scattering in the platinum foil backings, was determined by Monte-Carlo techniques. Scatter from the containment vessel used the plutonium measurements increased the flux at the target by (0.54 ± 0.14)%. Of lesser magnitude was the correction for scatter in the structure (0.33 ± 0.17)% and (0.42 ± 0.11)% for uranium and plutonium, respectively.

The calculation of the solid angle detection efficiency accounted for the anisotropy of fission fragment emission, the measurement thickness distribution, and the scalar flux variation over the deposit area. Incorporated in the code was the functional form of the fragment emission angular dependence, $W(E,\theta) = 1 + A(E) \cos^2\theta$, where θ is the angle between the incident neutron direction and the fragment emission in laboratory coordinates. Experimental data for $A(E)$ from Cavana (1974), Hsue (1976), Leachman (1965), Nesterov (1966), Nadkarni (1968), Simmons (1960), Smirenkin (1970), Androsenko (1971) and Shpak (1971) were first weighted, then averaged over the Cf neutron spectrum. Although the variance in the data is significant between 0.5–1.5 MeV, the spectrum-averaged value for $A(E)$ can be determined with more certainty than the differential data over this range. For both ^{235}U and ^{239}Pu, the spectrum averaging yielded $\bar{A} = (0.160 \pm 0.030)\%$ leading to uncertainties of 1.28% and 1.36% in their respective cross-sections.

The correct normalization of the $W(E,\theta)$ function must account for the momentum of the compound nucleus and the slight subsequent forward bias in the angular distribution. The emission in the forward 2π solid angle was found to be 1.0073 ± 0.0018 and 1.0072 ± 0.0018 fragments per fission for ^{235}U and ^{239}Pu, respectively, by means of simple kinematic calculations based on average fragment mass values.

In addition to the imprecision always associated with Poisson distributed counts, an uncertainty of 0.5% due to scanner bias was assumed based on the average difference in the repeated sample counts. A registration efficiency of 100% ± 0% has been assumed for the track-etch method of fission counting using solid angle counting on the basis of tests reported by Gilliam and Knoll (1970).

3.2. Neutron source calibration corrections

The determination of the neutron source strength from the manganese bath data involves several adjustments and corrections. The manganese bath mixing behavior has been sufficiently well determined from prior work (Robertson et al., 1975) so that the adjusted saturated activity values showed no deviation other than statistical variation over the entire 36-hour activation–decay cycle.

The parasitic neutron absorption was approximated by the products of the measured re-entry flux and the total macroscopic thermal absorption cross-section of all nuclei in the source and dry well. A parasitic absorption value of (0.12 ± 0.02)% and (0.71 ± 0.12)% for Cf and NBS-II, respectively, led to the total correction of (0–0.59 ± 0.13)% in the source comparison.

The neutron streaming from dry well was calculated from the effective solid angle of the opening with consideration given to streaming of the slow neutrons returning to the cavity as well as the direct streaming from the source. ANISN transport calculations of the bulk penetration leakage were verified by long counter measurements. The correction for leakage in the source comparison was determined to be (0.22 ± 0.05)%.

Table 1. Comparison of fast capture loss correction to manganese bath for Cf fission spectrum

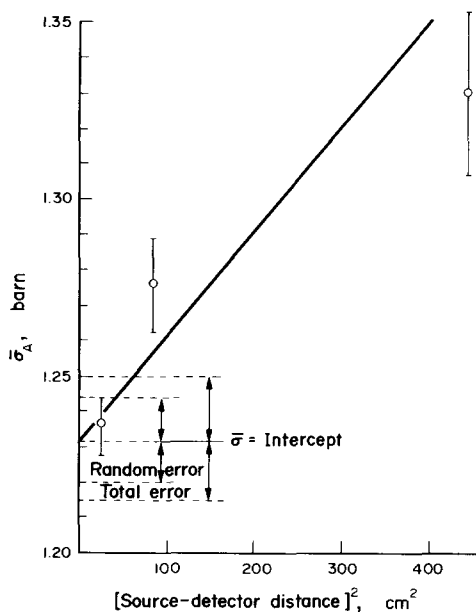| Calculation	Mn concentration C_H/C_{Mn}	g/l	% of neutron capture oxygen	sulfur	total
Engdahl (1976)	45	1.31	0.472	0.170	0.642
Ryves et al. (1965)	30		0.43	0.28	0.68 ± 0.10
Axton (1976)	30				0.80 ± 0.10
McTaggert (1961)		1.465	0.34 ± 0.05	0.20 ± 0.01	0.54 ± 0.05
Murphy (1965)		1.00			0.61
		1.219			0.76
		1.300			0.86
		1.465			0.96

Fig. 3. The apparent $^{235}U(n,f)$ integral cross-section over the ^{252}Cf fission spectrum vs R_{eff}^2.

For neutron energies below 2 MeV, the manganese bath comparison is independent of the source energy. That is, the ratio of the source neutron captured by ^{55}Mn to those captured by hydrogen is constant. However, above 2 MeV other nuclei begin to compete for neutron capture. A correction to the bath efficiency had to be made for fast neutron losses due to reactions such as $^{16}O(n,\alpha)$ and $^{32}S(n,p)$. Fast neutrons are absorbed effectively by these reactions, never reach thermal neutron energies, and thus can never activate manganese atoms. Table 1 shows the results of calculations by Murphy (1965), Ryves and Harden (1965), McTaggert (1961) and Axton (1976). The value chosen was $(0.64 \pm 0.20)\%$ based on a ANISN transport calculation and where the error is that recommended by Casewell (1971).

3.3. The number of target nuclei

The masses of the U_3O_2 deposits were determined to be 8.16 mg and 6.25 mg by the Isotope Target Laboratory at ORNL. The uncertainty was taken as 0.50% for reasons discussed by Gilliam and Knoll (1975). The plutonium deposit was alpha counted relative to the NBS foil standard, 49K-4-1, whose isotopic composition was identical to the foils used in these measurements. Masses of 5.59 mg. and 4.74 mg. $\pm$ 1.4% were determined.

A correction to account for the isotopic fission contribution to the total fission rate was $(-0.40 \pm 0.12)\%$ and $(-0.02 \pm 0.04)\%$ for the uranium and plutonium cross-sections, respectively.

3.4. Apparent cross-section

The total neutron flux at the deposit is the sum of the direct streaming from the source and of the room-scattered flux. The room-scatter is proportional to the source strength just as is the direct flux. Contributors include the cement walls, containment drum, the emptied manganese bath system and any scatters not previously accounted for which the magnitude is independent of detector spacing. Hence, as seen in Fig. 3, the cross-section for direct source neutrons as determined by plotting the apparent cross-section at varying source–detector distances and extrapolating to zero distance.

4. RESULTS AND DISCUSSION

Values of 1.215 ± 0.022 barn for ^{235}U and 1.790 ± 0.041 barn for ^{239}Pu were obtained for the integral fission cross-section, corresponding to a ratio value of 1.473 ± 0.041. Tables 2 and 3 summarize the corrections and uncertainties, respectively. The sign attached to each correction is the sign of the associated change in the cross-section value due to the particular adjustment cited. The final quoted error is the

Table 2. Adjustments and corrections with residual errors

Type of perturbation	Resulting correction (%) ^{235}U	^{239}Pu
Fragment emission anisotropy	-7.00 ± 1.28	-7.83 ± 1.36
Angular distribution normalization to lab	-0.73 ± 0.18	-0.72 ± 0.18
Fast capture losses	-0.64 ± 0.20	-0.64 ± 0.20
Parasitic absorption	$+0.59 \pm 0.13$	$+0.59 \pm 0.13$
Leakage	-0.22 ± 0.05	-0.22 ± 0.05
Scattering in Pt backings	-1.38 ± 0.17	-1.38 ± 0.17
Scattering in other structures	-0.42 ± 0.17	-1.05 ± 0.18
Foil impurities	-0.40 ± 0.12	-0.02 ± 0.04

Table 4. Comparison of Cf spectrum-averaged cross-sections

Integral	Cross-section		^{239}Pu(n,f)/ ^{235}U(n,f)
	^{235}U(n,f)	^{239}Pu(n,f)	
This work	1.215 ± 0.022	1.790 ± 0.041	1.473 ± 0.041
Heaton	1.205 ± 0.027	1.808 ± 0.045	1.500 ± 0.024
ENDF/B-IV	1.241 ± 0.002[a]	1.789	1.442
ENDF/B-III	1.239	1.819	1.468
ENDF/B-V	1.233	—	—

Errors are those due only to the uncertainty in the Cf fission spectrum.

Table 3. Summary of major uncertainties

Experimental factor	Resulting uncertainty (%)	
	^{235}U	^{239}Pu
Propagated error in net fission cts/source neutron	0.74	0.75
Manganese bath comparison of sources	0.30	0.30
Fragment emission anisotropy	1.28	1.36
Angular distribution normalization to lab	0.18	0.18
NBS-II reference source	0.50	0.50
Foil masses	0.50	1.40
Scattering in Pt backing and structures	0.24	0.24
Propagated errors in compensated beam geometry	0.57	0.57
Total random error	0.95	0.96
Total systematic error	1.51	2.05
Quadrature sum	1.79	2.27

quadrature sum of the independent error components.

Table 4 gives a comparison of this work with that of Heaton *et al.* (1976) and various differential data weighted over the Cf spectrum. Spectrum-averaged measurements of this type can serve as a useful check on various data sets, particularly in terms of their normalization level in the 0.5 to 5.0 MeV range. Since many features of the present measurements (target foils, counting apertures, manganese baths, etc.) are in common with previous measurements at Michigan, they also support the validity of results reported earlier using photoneutron sources (Gilliam and Knoll, 1976; Davis *et al.*, 1976).

REFERENCES

Androsenko K. D. and Smirenkin G. N. (1971) *Sov. J. nucl. Phys.* **12** (2), 143.
Axton E. J. (1976) unpublished report.
Carvana J., Mathur J. N., Boldeman J. W. and Walsh R. L. (1974) *Proc. Indian Nucl. Phys. Solid State Phys. Sym.*, Trombay, 1974.
Caewell R. S. (1971) *Am. Nucl. Soc. Topical Meeting*, Augusta, Vol. 1, pp. 1–53.

Davis M. C., Knoll G. F., Robertson J. C. and Gilliam D. M. (1978) *Ann. nucl. Energy* **5**, 569.
Davis M. C., Knoll G. F. and Robertson J. C. (1976) *NEANDC/NEACRP Spec. Meeting on Fast Neutron Cross Sections*, Argonne, *NEANDC (US)*-199/L, p. 225.
Engdahl J. C. (1976) Master's dissertation, University of Michigan.
Engle W. W. Jr. (1967) ANISN, A one dimensional discrete ordinates transport code with anisotropic scattering, *K*-1693, Oak Ridge Gaseous Diffusion Plant.
Gilliam D. M. and Knoll C. F. (1975) *Ann. nucl. Energy* **2**, 637.
Gilliam D. M. and Knoll G. F. (1970) *Trans. Am. nucl. Soc.* **13**, 526.
Grundl J. A. and Eisenhauer C. M. (1975) Nucl. cross section and tech., NBS Spec. Publ. 425, 2/25, Washington, D.C., p. 250.
Heaton H. T. II, Gilliam D. M., Spiegal G., Eisenhauer C. and Grundl J. A. (1976) *NEANDC/NEACRP Spec. Meeting on Fast Neutron Cross Sections*, Argonne, *NEANDC (US)*-199L, p. 333.
Hsue S. T. (1976) Master's dissertation, University of Michigan.
Huizenger J. R. and Behkami A. N. (1968) *Phys. Rev.* **174**, 1539.
Leachman C. M. and Blumberb L. (1965) *Phys. Rev.* **137B**, 814.
McTaggert M. H. (1958) AWRE Report *NR/A1/59*.

McTaggert M. H. (1961) *Reactor Sci. Technol.* **14,** 212.

Murphy W. M. (1965) *Nucl. Instrum. Meth.* **37,** 13.

Nadkarni D. M., Kapoor S. S. and Rama Rao P. N. (1968) *Nucl. Phys. State Phys. Sym.*, Bombay.

Nesterov V. G., Smirenkin G. N. and Shpak D. L. (1966) *Yad. Fiz.* **4** 933.

Robertson J. C., Davis M. C. and Engdahl J. C. (1975) Nuclear cross section and tech., NBS Spec. Publ. 425, Washington, D.C., p. 112.

Ryves T. B. and Harden D. (1965) *J. nucl. Energy* A/B **19,** 607.

Shpak D. L., Ostapenko Yu. B. and Smirenkin G. N. (1971) *Yad. Fiz.* **13,** 950.

Simmons J. E. and Henkel R. L. (1960) *Phys. Rev.* **120,** 198.

Smirenkin G. N., Shpak D. L., Ostapenko Yu. B. and Fursov B. I. (1970) *J. exp. theor. Phys. Lett.* **11,** 333.