Nuclear Data for Science and Technology (1988 MITO), 1033-1036, Copyright © 1988 JAERI.

Cross-Section Measurements in the Intermediate-Energy Standard Neutron Field

G.P. Lamaze, C.M. Eisenhauer, J.A. Grundl,
E.D. McGarry, F.J. Schima, and V. Spiegel

National Bureau of Standards, Gaithersburg, MD, 20899, USA

Abstract: Spectrum-averaged cross sections of six reactions of interest to neutron dosimetry have been measured in the Intermediate Energy Standard Neutron Field (ISNF) at the National Bureau of Standards (NBS). The ISNF neutron energy spectrum resembles a fast reactor spectrum, but has a much less complex structure due to the simplicity of its spherical configuration. The median energy is about 0.58 MeV, with 98% of the neutron fluence between 1.2 keV and 5.6 MeV. The ISNF neutron fluence is determined by fluence transfer from a calibrated ^{252}Cf fission source. Samples of ^{235}U were irradiated to a known fluence in compensated beam geometry in the ^{252}Cf field and the ^{140}Ba-^{140}La count rate determined with a high purity germanium detector. Similar ^{235}U samples were then irradiated in the ISNF as a fluence monitor and counted in the identical geometry. The following spectrum-averaged cross sections were obtained: ^{23}Na(n,γ)^{24}Na = 1.57 ± .10 mb, ^{45}Sc(n,γ)^{46}Sc = 24.4 ± 0.8 mb, ^{59}Co(n,γ)^{60}Co = 36.3 ± 1.5 mb, ^{109}Ag(n,γ)^{110m}Ag = 23.8 ± 0.9 mb, ^{115}In(n,n')^{115m}In = 97.0 ± 2.5 mb, and ^{197}Au(n,γ)^{198}Au = 411 ± 11 mb. Comparisons with calculated spectrum-averaged cross-sections are presented.

(fast neutrons, cross section, ^{115}In(n,n')^{115m}In, ^{197}Au(n,γ)^{198}Au, ^{23}Na(n,γ)^{24}Na, ^{59}Co(n,γ)^{60}Co, ^{109}Ag(n,γ)^{110m}Ag, ^{45}Sc(n,γ)^{46}Sc)

Introduction

The Intermediate-Energy Standard Neutron Field (ISNF) is an irradiation facility designed to produce a strong component of neutrons in the energy range of interest for fast-reactors and related technologies. This facility has been described[1-5] in detail elsewhere. Briefly, the ISNF consists of eight ^{235}U disks symmetrically placed near the surface of a 30 cm diameter spherical cavity in the graphite thermal column of the 20 MW Research Reactor at the National Bureau of Standards (NBS) in Gaithersburg, MD. The ^{235}U disks provide a source of fission spectrum neutrons for the irradiation field. An inner spherical shell of ^{10}B (14.4 cm outside diameter and 1.29 cm thick) provides a 1/v absorber to remove thermal neutrons and the lower energy part of the moderated spectrum of neutrons reflected from the graphite thermal column. Samples to be irradiated are placed near the center of the ^{10}B shell. A reentrant hole is available for active instrumentation or the shell can be completely sealed for passive irradiations. A schematic of the facility is shown in the top view of Figure 1.

The spherical symmetry of the system, together with the use of materials whose nuclear cross sections are among the best known, allow the resulting spectrum shape to be calculated with some confidence. The spectrum is limited in accuracy only by the uncertainty in the driving spectrum and in the input cross-section data of the materials making up the ISNF. Such calculations, employing both discrete-ordinates and Monte-Carlo methods, have been carried out by NBS[1], Oak Ridge[5], and Los Alamos[6,7]. The simplicity of a one dimensional calculation permits detailed investigation of spectrum and cross-section sensitivity to uncertainties in physical and nuclear parameters of the system and spectrum perturbation due to extraneous material required for fabrication. Figure 1 (lower) shows the ISNF spectrum together with the starting fission spectrum and the spectrum obtained without the ^{10}B shell. The low-energy tail below 10 keV is calculable to better than 5%. By measuring spectrum-averaged cross sections of activation dosimeters in this well-characterized field, a validation of the differential cross sections can be obtained.

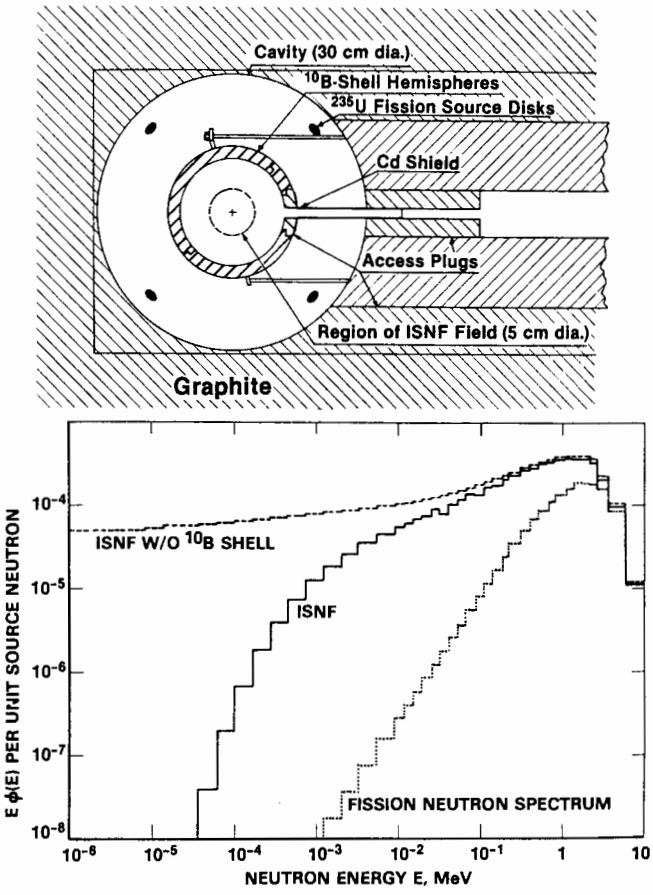

Figure 1. Upper view: The Intermediate-energy Standard Neutron Field located within a 30 cm diameter cavity in the thermal column of the NBS reactor. Lower view: Comparison of the ^{235}U fission spectrum to the ISNF spectra with and without the ^{10}B filter.

Experimental Procedure

While the energy dependence of the fluence comes from transport calculations, the magnitude of the neutron fluence is obtained through a procedure known as fluence transfer. At NBS, this begins with NBS-I, the national standard radium-beryllium photoneutron source. The most recent calibrations of this source were made in 1961[8] and 1978[9] and the calibrations agree to within 0.25%. This source is used to calibrate more intense working sources of ^{252}Cf. The 4π neutron emission rates of the ^{252}Cf spontaneous fission sources are determined in a 1.2 m diameter manganous sulfate bath by comparing induced ^{56}Mn activities with that produced by NBS-I. This technique is capable of yielding a ^{252}Cf source strength with an uncertainty of ± 1.1% (1σ).

The ^{252}Cf sources are thinly encapsulated in a light-weight aluminum and stainless steel capsule. The source can be suspended 1.6 m from the nearest reflecting surface in a low-scatter environment producing a nearly pure fission spectrum whose field strength depends only on the source emission rate and source-to-detector distance. This field can then be used to measure spectrum-averaged cross sections ($\bar{\sigma}_{Cf}$) directly or, through the process of fluence transfer, be used to determine the fluence rate in another field.

For the ISNF, the ^{235}U(n,f)^{140}Ba-^{140}La reaction was selected as the transfer reaction. Two thin uranium foils enriched to 93% in ^{235}U were wrapped in aluminum foil and placed on opposite sides of the Cf source and at an average distance of 4.7 cm from the source. In this compensated beam geometry, the first-order distance uncertainty is associated only with the separation of detectors, which can be measured to ± 0.01 mm. The more imprecise positioning of the source introduces only a second order uncertainty. The uranium foils were irradiated for a total of 20 days (66.8% of saturation) at the ^{252}Cf irradiation facility. After the ^{140}Ba and ^{140}La were in equilibrium, the sample activity was counted in a fixed geometry (through 0.6 cm of lead to remove most of the low energy activity) with a 23% efficient intrinsic germanium detector. The absolute efficiency of this counting system is not important, only the counting geometry must be reproducible and the efficiency remain constant. The gamma detector is then effectively calibrated in terms of ^{252}Cf fluence rate per detector counting rate (n cm^{-2}s^{-1}/counts s^{-1}g^{-1}).

When ^{235}U is used to monitor an ISNF irradiation, the calibration must be adjusted to account for the difference of the spectrum-averaged cross sections of ^{235}U in the two fields. The adjustment factor is the calculated spectrum-averaged cross-section ratio:

$$\bar{\sigma}_{Cf}/\bar{\sigma}_{ISNF} = 1.236/1.602 = 0.772 \pm 1.6\%.$$

(A further correction is made for the presence of ^{238}U in the samples, which alters the calculated cross-section ratio by 1.2%.) The uncertainty in this adjustment factor is related to the more general issue of ISNF fluence transfer options. This matter is discussed in Part IIA of reference 2. The uncertainty components for the cross-section ratio in the ^{235}U fluence transfer option are listed in Table I.

TABLE I

Uncertainty Components
in ^{235}U Cross-Section Ratio

^{235}U cross-section shape[11]	1.5 %
ISNF spectrum[2]	0.6 %
Cf fission spectrum[2]	0.1 %
Total Uncertainty	1.6 %

It should be noted that the fluence transfer option employed here, ^{235}U(n,f), establishes a fluence scale that is approximately 2% higher than the ^{239}Pu(n,f) transfer used for fission cross sections reported by Grundl and Gilliam.[10] The cross-section results reported there should be adjusted downward by 2% to be consistent with a ^{235}U fluence transfer.

The sample holder for the sealed configuration of the ISNF is shown in Figure 2. Two different thickness ^{235}U samples are placed in the sample holder near the center of the ISNF. Thin foils of gold, cobalt, indium, silver, scandium and sodium chloride (either pressed pellets or single crystals of salt) were also placed in the holder. The entire assembly is placed in the cavity of the graphite thermal column and the irradiation is begun by raising a boral curtain to allow reactor neutrons to enter the thermal column. The variations in the neutron intensity is monitored by a fission chamber placed in a nearby beam from the thermal column. Corrections for reactor power variations were less than 0.5%. The irradiation is terminated by lowering the boral curtain and removing the samples after a short cooling period. Two separate irradiations were made, each lasting 12 hours. A third irradiation of 18 hours was made without the fission drivers to measure the effect of background neutrons from the reactor core and the graphite.

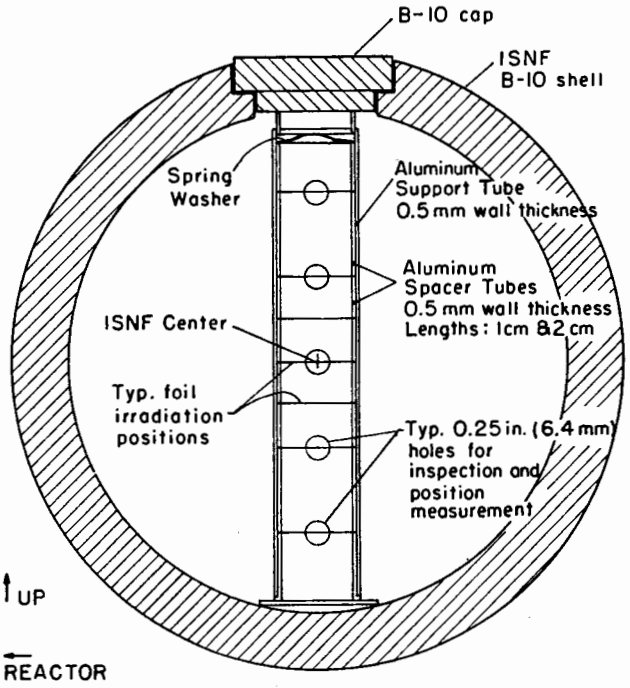

Figure 2. Sample holder for the sealed configuration of the ISNF. All samples were located within 1.2 cm of the center.

The neutron induced radioactivities were determined by measuring gamma-ray emissions with either of two efficiency-calibrated intrinsic germanium detectors. The primary detector is an 11.7% equivalent efficient detector with an energy resolution at 1332 keV of 1.2 keV FWHM. The samples were placed at 25 cm from the detector face. A computer-based data acquisition system was programmed to make periodic evaluation of the pre-selected gamma-ray emission rates. The peak fitting procedure is described in a report by F.J. Schima et al.[12] Data of each run were adjusted for decay to the time at the end of the neutron irradiation. Small corrections were made to the emission rates for the finite size and for the self-attenuation of the gamma-rays in each foil. A secondary detector of 23% equivalent efficiency was also used to measure some of the low-activity samples. The efficiency of this detector was calibrated relative to the primary detector or directly with standard calibration sources. Samples were placed at a distance of 3.7 cm from the front face of this detector.

Results

Indium

The $^{115}In(n,n')In^{115m}$ reaction is the only threshold reaction investigated in this set of irradiations. Four 0.25 mm thick and 12.7 mm diameter natural indium foils were irradiated in the first irradiation and three in the second. The background run indicated a 0.75% contribution of background neutrons which was subtracted. The correction for self-absorption of the 336 keV gamma rays in the foil was 1.2%. The cross section obtained in the two separate runs differed by 0.3%. Using a half-life of 4.486 h, a branching ratio of 0.458 and a natural abundance of .957, a spectrum-averaged cross section in the ISNF of 97.0 ± 2.5 mb was obtained. A value of 91.7 mb ± 3% can be inferred from an $^{115}In(n,n')/^{235}U(n,f)$ ratio measurement in the ISNF by Fabry et al[13]. A calculated value[2] using ENDF/B-V differential cross sections gives a value of 86.6 mb.

Gold

This reaction in the ISNF was investigated extensively by Fabry et al[13] and this work borrows heavily from some of their findings. Because of the strong gold resonances from 0.25 to 5 kev, self-absorption effects must be taken into account. Fabry et al[13] made measurements with seven different foil thicknesses to measure this effect. In the present measurement, two different thicknesses of pure gold samples and one sample of a gold-aluminum alloy were used. Sample thicknesses ranged from 1.3 mg/cm² for the alloy to 50 mg/cm² for the thickest pure gold foil. The Fabry[13] data was fit with a polynomial equation which was used to correct the present data to zero thickness. The corrections ranged from 0.1% for the alloy to 4.1 % for the 50mg/cm² sample.

An energy-dependent fluence gradient exists for low-energy neutrons along the vertical axis of the ISNF and the present data have been corrected for this effect using some of the results of reference 13. Those results indicated that there was both a symmetric and an asymmetric term, the latter due to asymmetries in the fission driver intensities. Since there is no way of knowing if the present fission drivers have the same asymmetry, only the symmetric term was used to correct the present data. The largest gradient correction was 1.4%.

After correcting the data for these two effects, and subtracting 0.6% for the background, an average value for the two runs of 411 ± 11 mb was obtained (the two runs differed from each other by one per cent). A value of 398 mb ± 3% can be inferred by the ratio measurement of Fabry et al.[13] A calculated value[2] of 392 mb has been obtained using ENDF-B/V differential cross sections.

Sodium

The sodium samples for the first irradiation were reagent-grade salt that were pressed into 12.7 mm diameter pellets that were 1.3 mm thick. No effort was made to dry the salt before or after pressing. To measure neutron self absorption, two pellets were stacked together and wrapped in aluminum foil. For the second irradiation, single crystal salt "windows" were obtained from a commercial source. These crystals were 13 mm in diameter and 2 mm thick. A single sample and a double sample were wrapped in aluminum foil for the irradiation. The background subtraction was about 0.6%. The extrapolation to zero thickness to correct for resonance self-absorption varied from 4% for the thinnest pressed samples to 12% for the single crystal pellets. The fluence gradient for the $Na(n,\gamma)$ reaction has not been measured and no correction has been applied. If the gradient was as large as for the gold reaction, the correction would increase the final result by about 2%. The results of the two separate irradiations differed by about 7% with the single crystal result being higher. Since the actual salt content of the pellet is in some doubt, those data have been discounted and only the data from the single crystal irradiation have been used to obtain a cross-section value of 1.57 ± 0.10 mb. Because of the thickness of the samples and the unmeasured gradient, this result should be considered preliminary until a remeasure can be made with thinner samples. No previous measurement of this reaction is available, but a calculation[2] with ENDF/B-V differential cross sections yields 1.89 mb.

Scandium

The scandium samples were commercially obtained pure metal foils that were 12.7 mm in diameter and 0.15 mm thick. To measure neutron self absorption, two foils were stacked together. The background subtraction was about 0.4%. Count rates for the 889 keV line was about 0.2 c/s in the photopeak in the high efficiency detector. The extrapolation to zero thickness was 1.6%. The fluence gradient for the $Sc(n,\gamma)$ reaction has not been measured and no correction has been applied. If the gradient was as large as for the gold reaction, the correction would increase the final result by about 0.2%. The results of the two separate irradiations differed by about 1%. The final value of the cross-section is 24.4 ± 0.8 mb. No previous measurement of this reaction is available, but a calculation[2] with ENDF/B-V differential cross sections yields 27.1 mb.

Silver

The silver samples were commercially obtained pure metal foils that were 12.7 mm in diameter and 0.13 mm or 0.25 mm thick. The background subtraction was about 0.4%. Photopeak counting rates for the 659 keV and 885 keV line were about 0.2 c/s and 0.1 c/s, respectively. The extrapolation to zero thickness was 1.2%. The fluence gradient for the $^{109}Ag(n,\gamma)^{110m}Ag$ reaction has not been measured and no correction has been applied. If

the gradient was the same as for the gold reaction, the correction would increase the final result by about 0.2%. The results of the two separate irradiations differed by about 1.5%. The final value of the cross-section is 23.8 ± 0.9 mb. No previous result for this reaction is available, and no calculation of the integral cross section has been made to date because the differential cross section is not in the ENDF dosimetry file.

Cobalt

The cobalt samples were commercially obtained pure metal foils that were 12.7 mm in diameter and 0.06 mm or 0.25 mm thick. The reactor background subtraction was about 0.7%. The extrapolation to zero thickness was obtained by irradiating two 0.06 mm foils back-to-back and by irradiating a 0.25 mm foil. The extrapolation to zero was obtained by fitting the three different thicknesses with a quadratic equation. The correction from the thinnest foil to zero was about 12%. This indicates that in a future repeat of this experiment, a dilute foil should be used to reduce the uncertainty arising from the extrapolation to zero. However, the photopeak counting rate for the thin foils was only about .03 c/s in the high efficiency detector. The fluence gradient for the $Co(n,\gamma)$ reaction has not been measured but since the gold and cobalt have a similar single resonance low-energy response in the ISNF the same correction has been applied as measured for the gold reaction. The gradient correction ranged from 0 to 0.4%. The results of the two separate irradiations differed by about 2%. The final value of the cross-section is 36.3 ± 1.5 mb. No previous result for this reaction is available, but a calculation[2] with ENDF/B-V differential cross sections yields 40.9 mb.

TABLE II -- UNCERTAINTY SUMMARY

FLUENCE TRANSFER	
Source strength of Cf	1.1 %
Distance Measurement	0.6
U235 Cross-Section Ratio	1.6
U235 Scattering	0.6
Statistics	0.9
Total Fluence Uncertainty	2.3 %
ACTIVATION (TYPICAL)	
Fluence Gradients	1.0 %
Extrapolation to zero thickness	1.0
Neutron Background	0.3
Gamma Counter Efficiency	1.0
Statistics	0.6
Cross-Section Uncertainty (including fluence uncert.)	3.0 %

Discussion

Uncertainties

Table II summarizes the uncertainties in this experiment. The first part of the table gives the uncertainty in the measured fluence at the center of the ISNF and is therefore common to all of the measured cross sections. The second part of the table gives some typical values for the uncertainties in the measure of the activation. As has been mentioned, the extrapolation to zero for cobalt and sodium is larger than for the other reactions and the assigned uncertainties for these extrapolation terms is 3% for cobalt and 6% for sodium.

Summary

Table III provides a summary of the results obtained in this work and compares them with those of Fabry et al[13] for gold and indium. Comparisons are also given with the calculated integral cross sections.[2] The calculations use the Oak Ridge calculated ISNF spectrum and differential cross sections from ENDF/B-V. It is planned to add the silver calculation to this set soon. The last column gives the calculated to experimental (C/E) ratios.

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TABLE III -- MEASURED AND CALCULATED CROSS SECTIONS

	This Work (mb)	Fabry et al[13] (mb)	Calculated[2] (mb)	C/E
In(n,n')	97.0 ± 2.5	91.7 ± 2.8	86.6	0.89
Au(n,γ)	411 ± 11	398 ± 12	392.	0.95
Sc(n,γ)	24.4 ± 0.8		27.1	1.11
Na(n,γ)	1.57 ± 0.10		1.89	1.20
Co(n,γ)	36.3 ± 1.5		40.9	1.13
Ag(n,γ)	23.8 ± 0.9			