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NEODYMIUM, SAMARIUM AND EUROPIUM CAPTURE CROSS-SECTION ADJUSTMENTS BASED ON EBR-II INTEGRAL MEASUREMENTS

by

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DISCLAIMER

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Abstract

Integral capture measurements have been made for high-enriched isotopes of neodymium, samarium and europium irradiated in a row 8 position of EBR-II with samples located both at mid-plane and in the axial reflector. Broad response, resonance, and threshold dosimeters were included to characterize the neutron spectra at the sample locations. The saturation reaction rates for the rare-earth samples were determined by post-irradiation mass-spectrometric analyses and for the dosimeter materials by the gamma-spectrometric method. The HEDL maximum-likelihood analysis code, FERRET, was used to make a "least-squares adjustment" of the ENDF/B-IV rare-earth cross sections based on the measured dosimeter and fission-product reaction rates. Preliminary results to date indicate a need for a significant upward adjustment of the capture cross sections for ^{143}Nd, ^{145}Nd, ^{147}Sm and ^{148}Sm.

Introduction

In recent years, integral data (capture reaction rates and reactivity worth measurements in fast-reactor fields) have played an important role in the evaluation of fission-product capture cross sections of importance to reactor technology, especially the development of fast reactor systems/1/. In the simplest evaluation application for isotopes with sparse or no measured differential data, integral measurements have been used to normalize capture cross sections based exclusively on nuclear model calculations. For isotopes with a more extensive base of measured differential data, integral measurements have been used to make integral tests of cross-section curves based on the differential measurements and nuclear model calculations. Such integral tests have been helpful to the evaluator in sorting out normalization problems between differential measurements. In a more sophisticated application, integral data obtained from measurements in different spectra have been used to adjust both multigroup and/or point-wise cross sections/1/. This latter application requires a realistic treatment of the uncertainties and correlations in the integral data and in the a-priori flux spectra and fission-product cross sections.

A significant fraction of the integral data used in the fission product cross-section evaluation process comprises reactivity worth measurements in the fast reactor spectra of the STEK cores/2/ and activation capture rates in the fast neutron field of the Coupled Fast Reactivity Measurements Facility (CFRMF) at the Idaho National Engineering Laboratory/3/. This paper presents the integral capture results for enriched isotopes of neodymium, samarium and europium irradiated in different spectra in the Experimental Breeder Reactor-II (EBR-II). The Nd and Sm cross sections are of importance to fission product poison effects in fast reactors and/or to the establishment of a reliable burnup monitor for fast reactor fuels. Cross sections for the Eu isotopes are needed in the evaluation of europium oxide as a control material. For most of the isotopes in the irradiation, some integral data exist as reactivity worths. Little, if any, integral capture data have been published. The EBR-II experiment differs significantly from experiments in the CFRMF and STEK facilities in terms of neutron spectrum characterization. The neutron fields in the latter two facilities are well characterized by means of neutronic calculations and active neutron dosimetry. Characterization of the neutron spectra in the EBR-II is dependent on the use of passive dosimeters (activation monitors).

Included in this paper are a brief description of the EBR-II irradiation experiment and a detailed presentation of the measured reaction rates for the rare-earth samples and for the neutron spectrum dosimeters. In addition, preliminary results of the application of the FERRET Code/4,5/ for spectrum unfolding and for the adjustment of ENDF/B-IV multigroup cross sections based on the measured integral data are presented.

EBR-II Irradiation Experiment

Irradiation Configuration

A detailed description of the irradiation experiment was presented earlier/6/. Only pertinent details will be given here. Shown in Figures 1 and 2 are the subassembly and axial loading patterns for this experiment. The irradiation package consisted of multiple samples (0.1 μg to 50 μg deposits on Ni or V foils) of the isotopically enriched isotopes shown in Figure 2 and dosimeter sets consisting of Co, Cu, Fe, Ni, Ti, Sc, ^{237}Np, ^{235}U, ^{238}U monitors. Two B-7 capsules provided the primary containment of the eight experiment capsules. Each experiment capsule contained up to five subcapsules each of which contained the sample or dosimetry materials.

Reaction-Rate Determination

Dosimeters. Saturation reaction rates for the dosimeters were determined by the radiometric technique/7,8/ using calibrated Ge(Li) spectrometers. Decay data for the analysis was taken from reference 8. The fission-rate determinations were based on the consensus fast reactor fission yields given in reference 9. Infinitely-dilute reaction rates for the dosimeters in each set for the irradiation are summarized in Table I. Accurate fission rates for ^{238}U, not given in the table, are difficult to obtain because a large correction is required to account for fission-product activity due to fission of the "grown-in" ^{239}Pu. Uncertainties in the reaction rates for the Co dosimeters reflect significant neutron self-shielding corrections (~factor of 2) required for these monitors.

Rare-Earth Samples. The saturation reaction rates summarized in Table II for the Nd, Sm and Eu samples are based on mass-spectrometric or gamma spectrometric measurements for the post-irradiation samples. For the Nd and Sm isotopes for which integral results are reported here, both the parent and the capture products are stable and the reaction rates are determined easily from mass spectrometer measurements of the (A+1)/A atom ratios for the samples/6/.

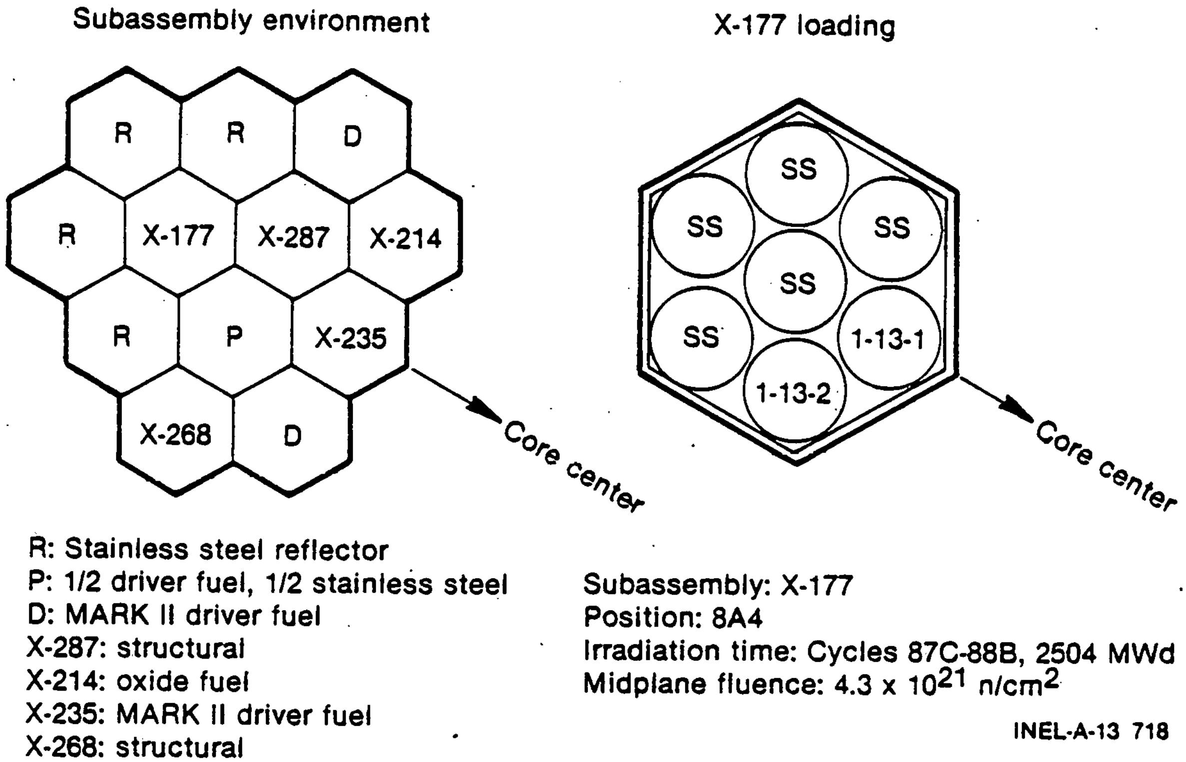

Fig. 1 Subassembly loading pattern for EBR-II Irradiation.

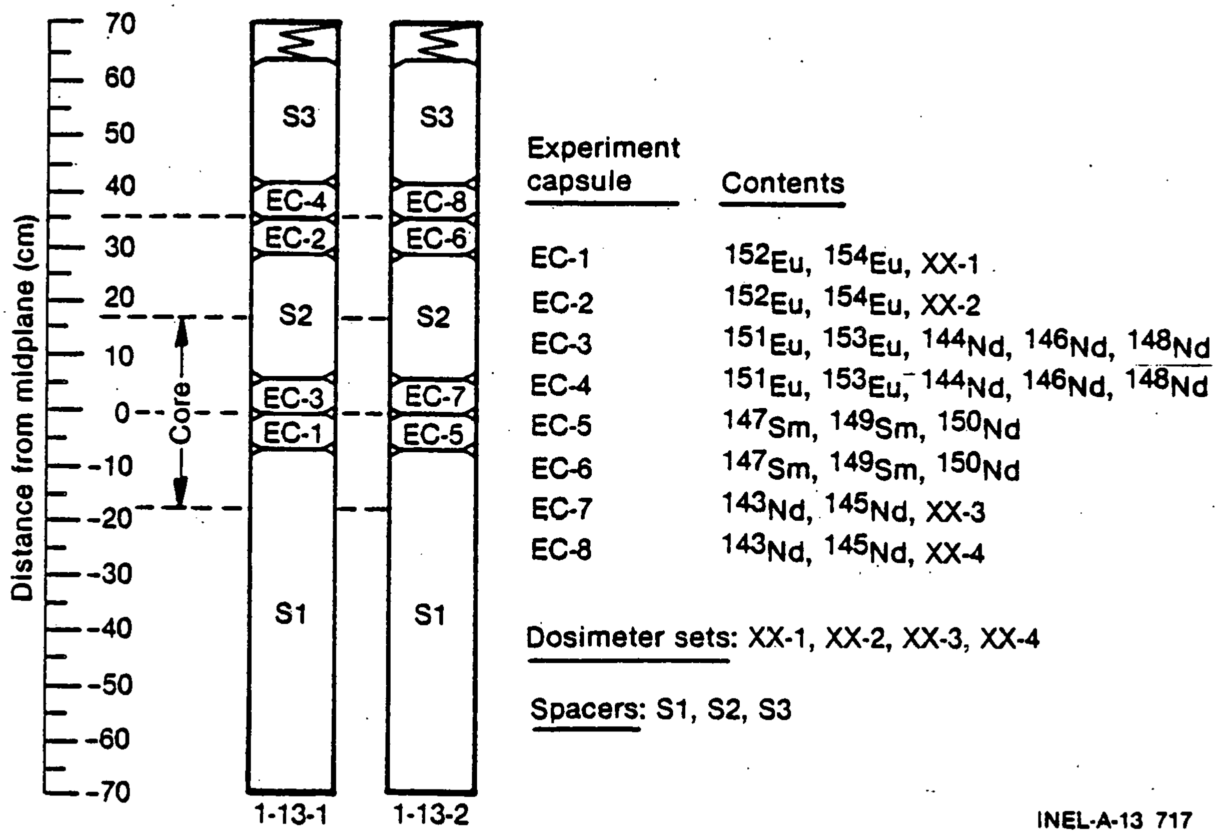

Fig. 2 B-7 Axial Loading Pattern for EBR-II Irradiation.

TABLE I. Infinitely-dilute Reaction Rates for Dosimeters in EBR-II Experiment X-177

Reaction	Reaction rate (reactions/sec-atom) x 10^{11} XX-1[a]	XX-3	XX-2	XX-4
$^{59}Co(n,\gamma)^{60}Co$	20.2(9)[b]	27.8(12)	64.(5)	70.(6)
$^{235}U(n,f)$	231.(7)	234.(9)	238.(9)	230.(8)
$^{237}Np(n,f)$	63.(4)	50.(2)	19.3(10)	11.0(5)
$^{45}Sc(n,\gamma)^{46}Sc$	3.75(9)	3.72(8)	3.99(9)	4.07(8)
$^{54}Fe(n,p)^{54}Mn$	1.300(22)	0.910(15)	0.2162(37)	0.1024(17)
$^{58}Fe(n,\gamma)^{59}Fe$	0.965(13)	0.998(14)	1.239(16)	1.166(15)
$^{58}Ni(n,p)^{58}Co$	1.74(4)	1.27(3)	0.306(6)	0.152(3)
$^{46}Ti(n,p)^{46}Sc$	0.1570(19)	0.1142(14)	.02364(28)	0.01169(14)
$^{63}Cu(n,\alpha)^{60}Co$	0.00728(12)	0.00527(8)	.00212(3)	0.00104(2)

[a]Label for dosimetry set.

[b]Number in parenthesis is the 1-sigma error in the last significant digits.

TABLE II. Infinitely-dilute (n,γ) Reaction Rates for Rare-Earth Samples in EBR-II Experiment X-177

Isotope	Applicable[a] Dosimeter Set	Reaction Rate (rps/atom)x10^{10}	$\frac{C}{M}$[c]
^{143}Nd	XX-3	5.03(6)[b]	.822
	XX-4	8.75(5)	.863
^{144}Nd	XX-1	1.06(2)	.997
	XX-4	0.993(11)	.970
^{145}Nd	XX-3	7.64(8)	.653
	XX-4	14.55(9)	.765
^{147}Sm	XX-3	23.11(14)	.658
	XX-2	44.8(5)	.813
^{149}Sm	XX-3	40.9(10)	.721
	XX-2	81.5(27)	1.07
^{151}Eu	XX-1	54.(3)	.705
	XX-4	113.(7)	.794
^{152}Eu	XX-1	52.(5)	.913
	XX-2	75.(8)	1.26
^{153}Eu	XX-1	29.6(15)	.804
	XX-4	61.9(25)	.934
^{154}Eu	XX-1	38.(3)	.757
	XX-2	62.(5)	1.03

[a]Dosimeter set identification which relates rare-earth reaction rates to dosimeter rates in Table I.

[b]Number in parenthesis is the 1-sigma error in the last significant digits.

[c]Calculated-to-measured reaction-rate ratios based on the unadjusted fission-product cross sections and the multigroup fluxes obtained from spectrum unfolding analysis.

Prior to mass spectrometric analysis, the rare-earth deposits were chemically isolated from the backing foil. A minimum of three mass-spectrometric analyses were made for the Nd and Sm samples from each axial location. The quoted errors for the Nd and Sm isotopes result from averaging the isotopic data from each mass-spectrometric analysis and accounting for an estimated 0.5% systematic error in the mass-spectrometric determination.

For the Eu isotopes, which involve radioactive parent or capture products, the reaction-rate determination is more complicated. Because a significant fraction of the capture in ^{151}Eu goes to the 9.6 h ^{152}Eu metastable state (estimated to be 41% from the data in reference 3), chemical isolation of the Eu fraction from the Gd and Sm decay products from the 9.6 h activity was required prior to mass-spectrometric analysis for the 152/151 atom ratio. Consequently, the capture rates for ^{151}Eu in the table were derived from decay-corrected measured-atom ratios divided by .59 to account for the isomer production. The errors in the measured capture rates for the ^{151}Eu are dominated by a 5% uncertainty estimated for the isomer ratio.

Capture rates for ^{152}Eu are based on decay corrected 153/152 atom ratios obtained from mass spectrometric measurements for Eu samples isolated from the nickel backing and from the Sm and Gd decay products from the decay of the 13.2y ^{152}Eu. The sizable errors estimated for the quoted ^{152}Eu capture rates result from uncertainties in the mass spectrometric determination of the 153/152 atom ratios in the unirradiated and irradiated samples. Similarly, the capture rates for ^{153}Eu are based on decay-corrected 154/153 atom ratios obtained from mass-spectrometric measurements. The dominant contribution to the error for the ^{153}Eu capture rate is due to uncertainties in the mass-spectrometric determination of the 154/153 atom ratios.

The capture rates for the ^{154}Eu samples are based on decay-corrected atom ratios determined by the Ge(Li) spectrometric measurement of the relative gamma emission rates of the 123.14-keV and 105.3-keV lines in the β^- decay of ^{154}Eu and ^{155}Eu, respectively. The dominant contributors to the uncertainty in the capture rates are errors in the gamma-ray branching ratios and half-lives used in the computation of the atom ratios from the relative gamma intensities. Decay data for these analyses were taken from the INEL Decay Data Master File .

Data Analysis

Neutron Spectrum Characterization

The FERRET data analysis code/4,5/, was used to obtain 47 group* representations of the neutron spectra based on the measured reaction rates for the dosimeters in Table I. A priori information for this analysis included the following:

1) 47 group fluxes derived from 29 group fluxes obtained from XY-geometry (for mid-plane) and RZ-geometry (for reflector) neutronics calculations for applicable core configurations of EBR-II/11/,

2) parametric representations for the flux covariance matrices,

3) 47 group dosimeter cross sections based on ENDF/B-IV, 620 group cross sections collapsed with a weighting function representative of the neutron spectra in EBR-II,

*Slightly modified version of the HEDL 42 group energy structure with maximum energy extended to 16.91 MeV.

4) parametric representations for the cross-section covariance matrices.

The covariance matrices generated for both the fluxes and cross sections are composed of two components: an overall fractional normalization uncertainty, c, and a second term, $r_i r_j \rho_{ij}$, that describes any additional uncertainties and correlations. The correlation matrix is parameterized by

$$\rho_{ij} = (1 - \Theta)\ \delta_{ij} + \Theta\ e^{-\frac{(i-j)^2}{2\gamma^2}}$$

where Θ denotes the strength of the short range correlations and γ denotes their range. For example, completely uncorrelated data or a-priori values are described by $\Theta=0$ so that $\rho_{ij} = \delta_{ij}$. The values, $\{r_i\}$ are the group-by-group fractional uncertainties.

In the present analysis, a mid-plane a-priori flux was assumed to have a 10% normalization uncertainty, a group-by-group uncertainty of 20% with short-range correlations specified by $\Theta = 0.9$ and $\gamma = 3.0$ A reflector a-priori flux was assumed to have a 20% normalization uncertainty and a group-by-group uncertainty of 40%. A more extensive evaluation by one of the authors (F. Schmittroth) of the uncertainties and correlations for the dosimeter cross section is beyond the scope of this paper.

Two examples of the spectrum-unfolding analysis which simultaneously treated all four dosimeter sets are illustrated in Figures 3 and 4. In Figure 3, one notes that the adjusted multi-group flux appears to be somewhat softer than the a-priori flux. Group-to-group fractional uncertainties were reduced

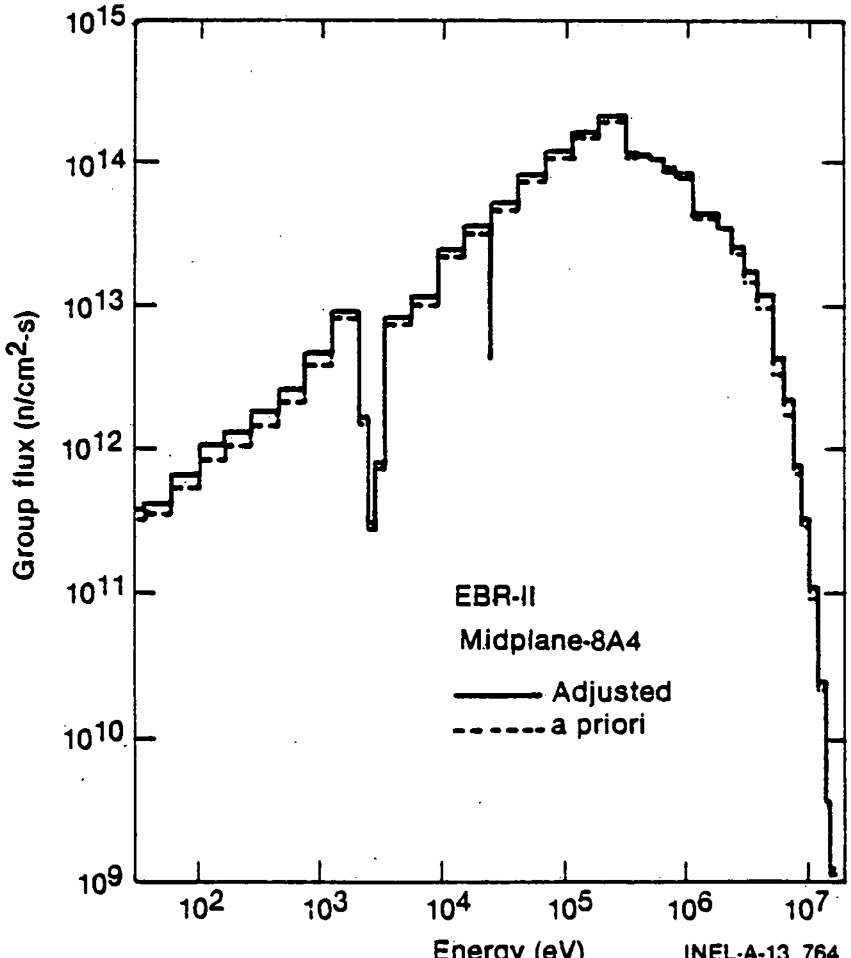

Fig. 3 Comparison of a-priori and adjusted multigroup fluxes for XX-1 dosimeter at mid-plane.

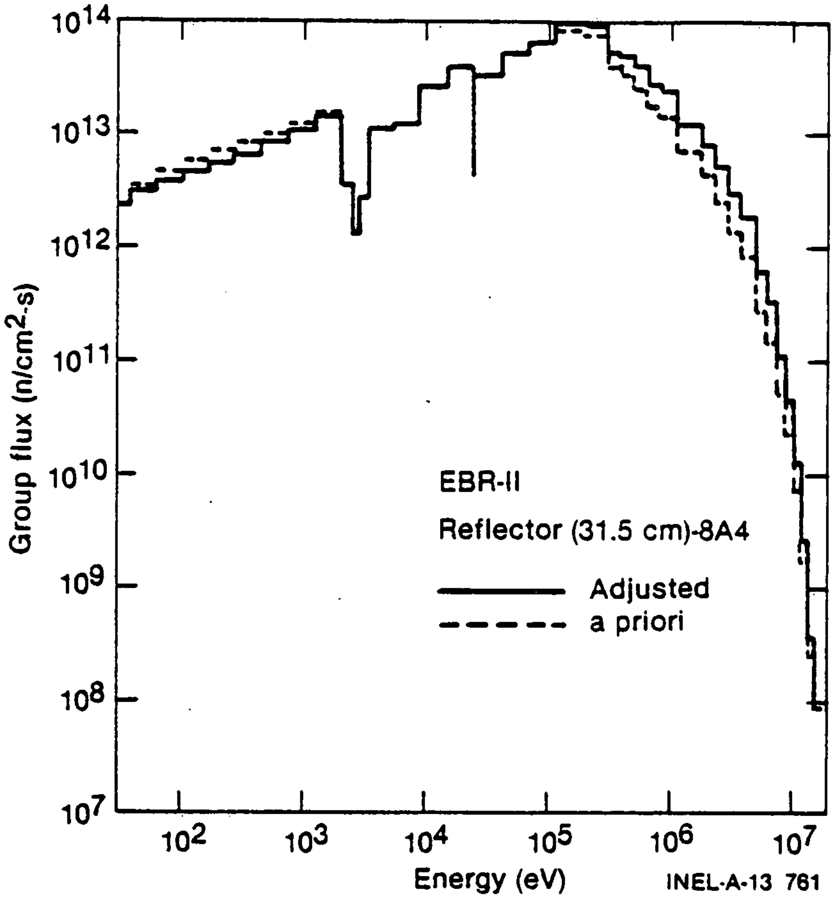

Fig. 4 Comparison of a-priori and adjusted multigroup fluxes for XX-2 dosimeter in the reflector.

to as low as 12% in the region of maximum response above the sodium dip (25 keV). Illustrated in Figure 4 is the overall hardening of the a-priori reflector neutron spectrum by the adjustment. The two figures illustrate the significant differences in the energy distribution of the neutron flux between a mid-plane and a reflector location and point to the sensitivity of the reflector reaction rates to resonance capture.

Cross-Section Adjustment

A least-squares adjustment of the fission-product multigroup cross sections was made with FERRET based on the following a-priori information:

1) Adjusted multigroup fluxes and adjusted flux covariance matrices from the spectrum unfolding analysis.

2) 47 group fission-product cross sections based on ENDF/B-IV.

3) parametric representations for the cross-section covariance matrices.

Summarized in the 4th column of Table II and illustrated by Figures 5-8 are some of the results of the FERRET analysis. The C/M ratios given in Table II present "conventional" integral tests of the fission-product cross sections. For example, both the mid-plane and reflector C/M ratios for ^{143}Nd indicate the need for an upward adjustment in the cross section throughout the region of sensitivity of the fluxes. The C/M ratios based on the adjusted fission-product cross sections and fluxes were essentially 1 for all cases except those with large errors in the measured reaction rate, e.g., ^{152}Eu.

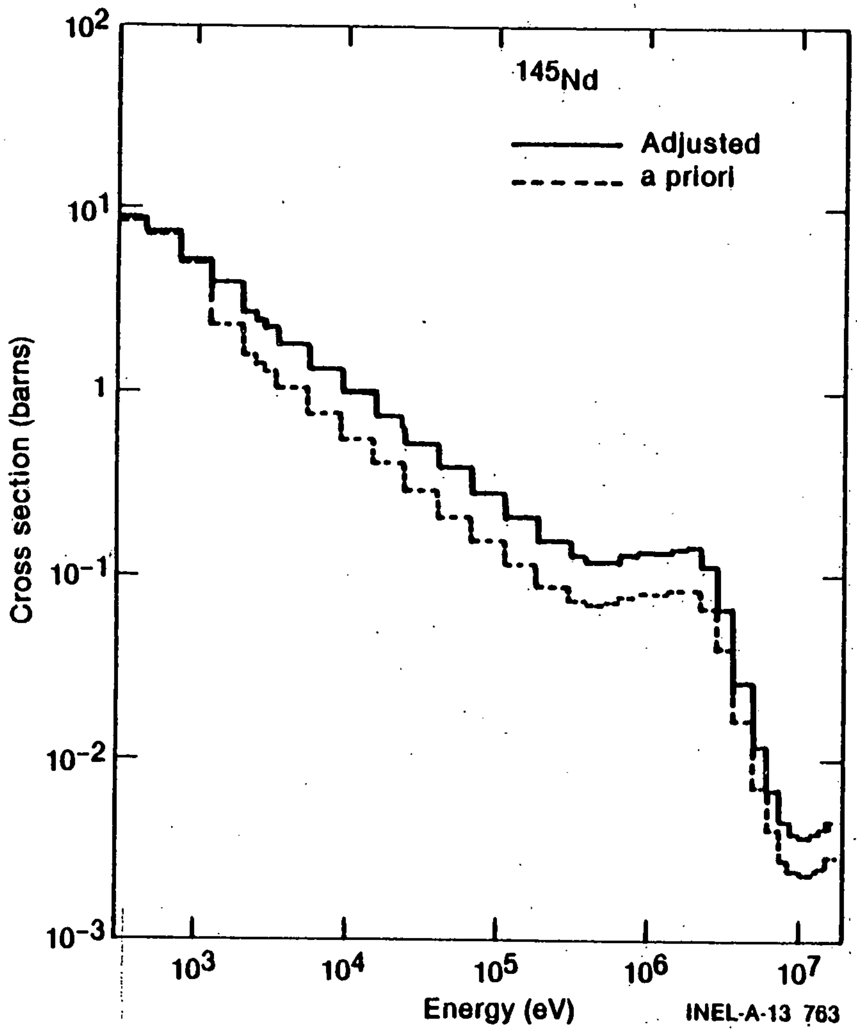

Fig. 6 Comparison of a-priori and adjusted multigroup cross sections for ^{145}Nd

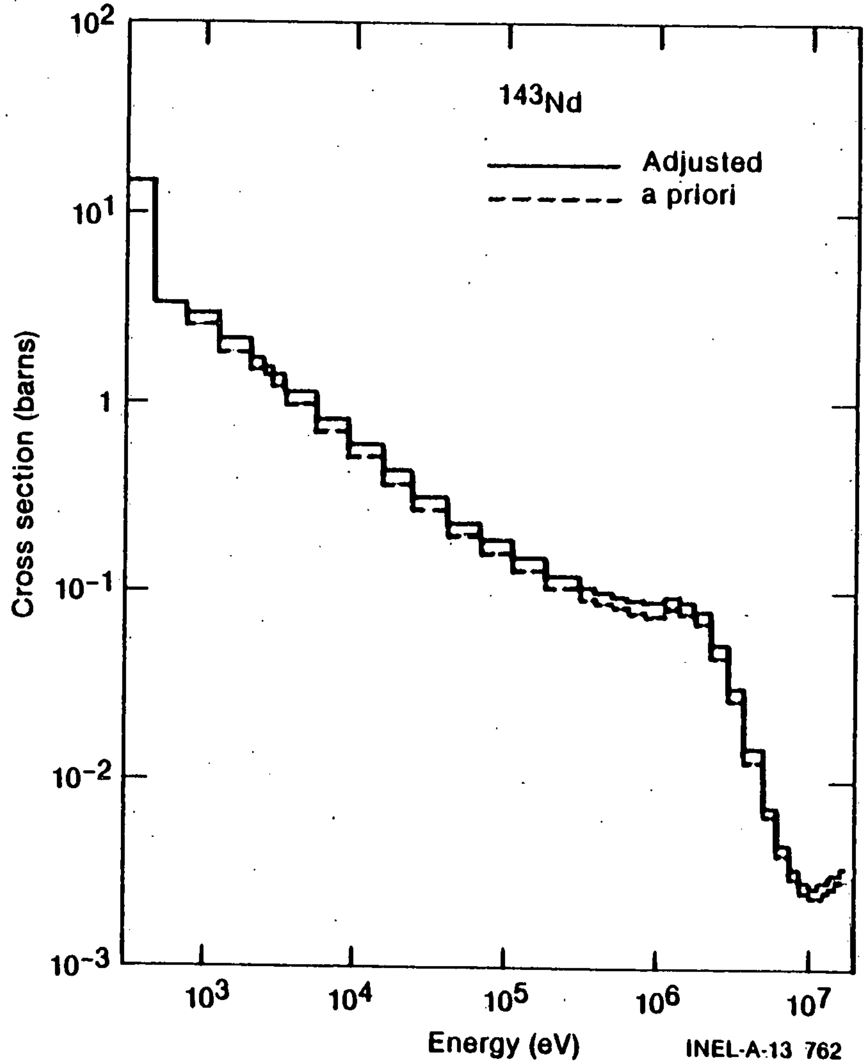

Fig. 5 Comparison of a-priori and adjusted multigroup cross sections for ^{143}Nd

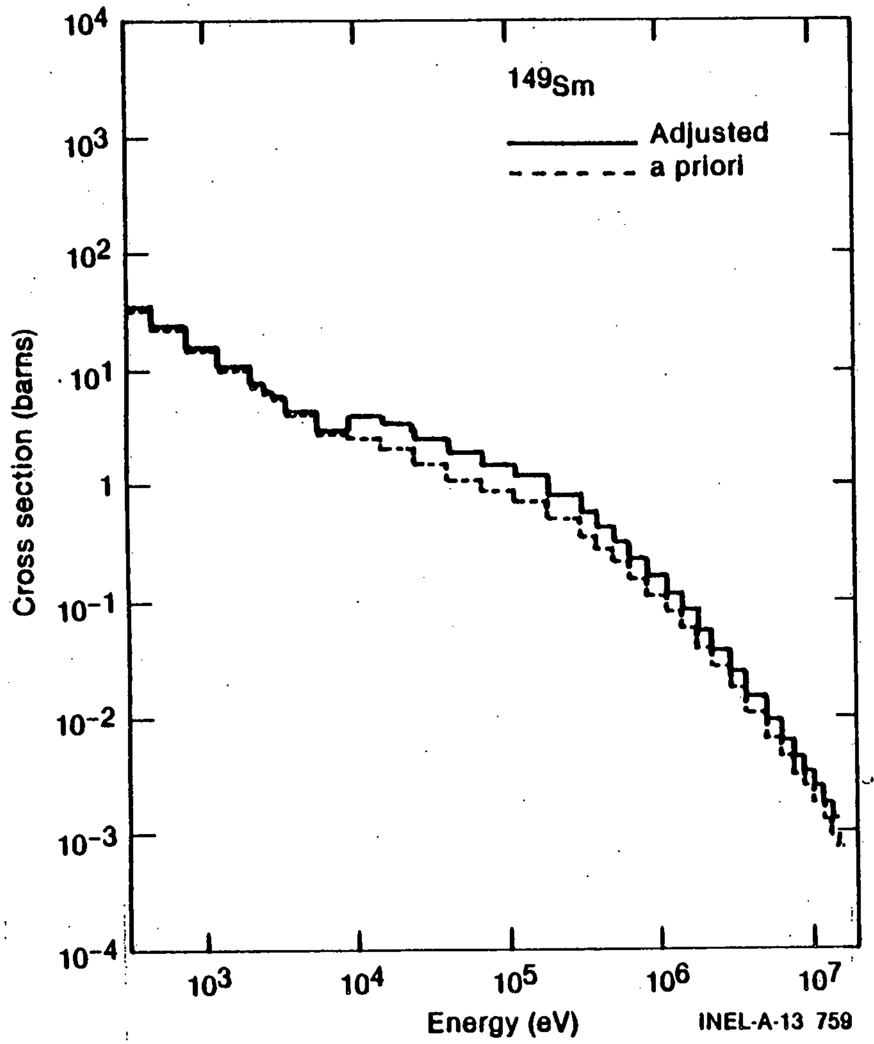

Fig. 8 Comparison of a-priori and adjusted cross sections for ^{149}Sm

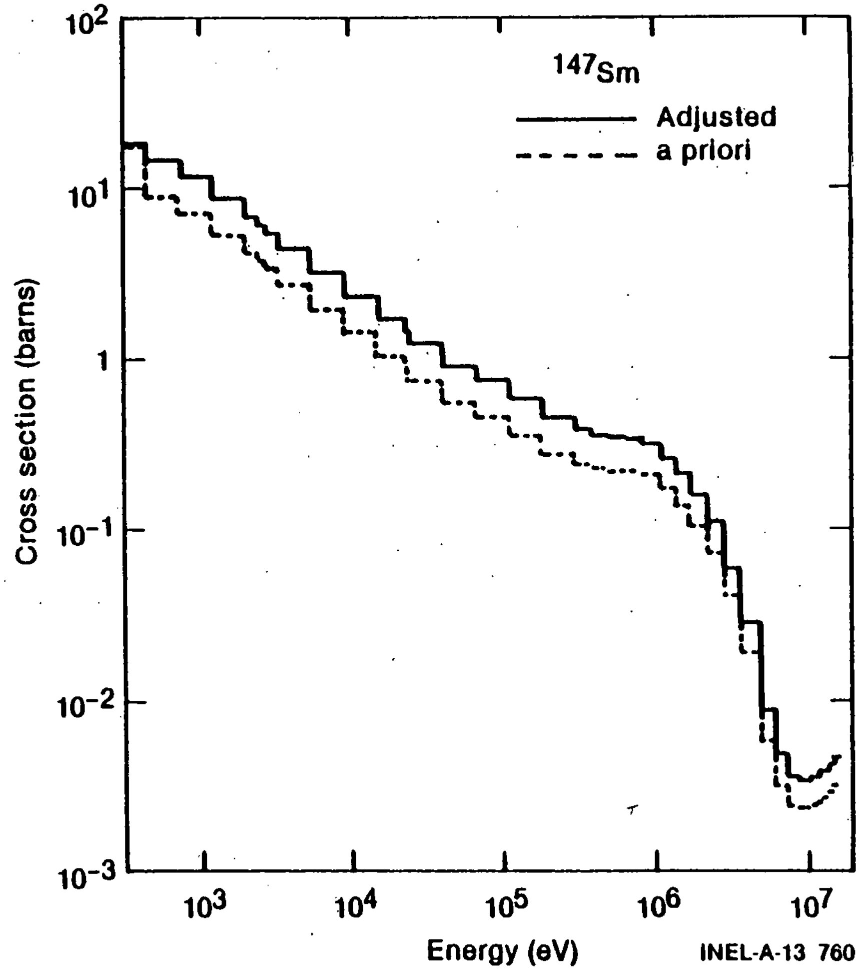

Fig. 7 Comparison of a-priori and adjusted cross sections for ^{147}Sm

Shown in Figures 5-8 are comparisons of the a-priori and adjusted cross sections for ^{143}Nd, ^{145}Nd, ^{147}Sm and ^{149}Sm isotopes for which no previous integral capture data have been reported. As expected, the adjustment in the cross section is mainly over the region of maximum response in the neutron fields and the magnitude of the adjustment is approximately given by the inverse of the C/M ratios from Table II.

Discussion

Some qualitative comparisons of the present integral results and cross section adjustments with other integral data, measured differential cross sections and/or evaluated cross sections were made. Integral checks of the ENDF/B-IV cross sections for ^{147}Sm and ^{149}Sm based on reactivity-worth measurements in the STEK cores has been reported as C/M ratios/2/. The C/M ratios from the present experiment are in good agreement with the STEK results. For ^{143}Nd, ^{144}Nd and ^{145}Nd, Gruppelaar/13/ has reported adjusted cross sections (RCN-2A set) based on reactivity-worth measurements in the STEK cores. A comparison of the adjusted Nd cross sections from the present work with those of Gruppelaar indicates good agreement for ^{145}Nd and reasonable agreement for ^{143}Nd and ^{144}Nd. A comparison of the preliminary EBR-II adjusted cross sections with recent differential data indicates reasonable agreement for ^{149}Sm and ^{145}Nd and good agreement for ^{143}Nd and ^{144}Nd/14/.

We consider the present integral results and FERRET analyses to be of more use in data evaluation that just for qualitative comparisons of C/M ratios and adjusted cross sections. Especially of use to the data evaluator are the adjusted cross sections and associated adjusted covariance matrices. The adjusted covariance matrices embody all the uncertainties and correlations associated with these integral experiments. The data could subsequently be used by the evaluator to adjust evaluated point cross sections based on nuclear model calculations and measured differential data/1/. Furthermore, with the advent and utilization of covariance files for ENDF/B cross sections, this approach to data evaluation will utilize in the most consistent way all the measured and calculated information important to determining point-wise cross sections for fission-product isotopes.

In summary, we would like to emphasize the unique features and contributions of the present experiment and analyses. From an experimental standpoint,

1) small sample sizes were required (μg quantities of highly enriched rare-earth samples were prepared with the INEL electromagnetic mass separator),

2) short-time (~30 days) irradiation of samples in different neutron fields of high flux test reactor,

3) spectral characterization of neutron fields by use of passive dosimetry,

4) capture reaction-rate measurements based on conventional mass spectrometric and Ge(Li) spectrometric techniques and capabilities at the INEL.

From an analysis standpoint, we have demonstrated the adjustment of cross sections based on the present integral results to be consistent with other evaluations and differential measurements.

Acknowledgment

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