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ACTINIDE INTEGRAL MEASUREMENTS IN THE CFRMF AND INTEGRAL TESTS FOR ENDF/B-V*

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Abstract

Integral capture and/or fission rates have been reported earlier for several actinides irradiated in the fast neutron field of the Coupled Fast Reactivity Measurements Facility (CFRMF). These nuclides include ^{232}Th, ^{233}U, ^{235}U, ^{238}U, ^{237}Np, ^{239}Pu, ^{240}Pu, ^{242}Pu, ^{241}Am and ^{243}Am. This paper focuses on the utilization of these integral data for testing the respective cross sections on ENDF/B-V. Integral cross sections derived from the measured reaction rates are tabulated. Results are presented for cross-section data testing which includes integral testing based on a comparison of calculated and measured integral cross sections and testing based on least-squares-adjustment analyses.

I. Introduction

Integral reaction rates have been measured for several actinides irradiated in the fast neutron field of the Coupled Fast Reactivity Measurements Facility (CFRMF)[1,2]. The purpose of this paper is to summarize this integral data base and to present the results of using these integral data to test the respective cross sections on ENDF/B-V. The impact of this work on the radiative neutron capture reactions is highlighted. Section II of this paper includes a brief description of the CFRMF, an identification of the types of integral measurements and a tabulation of integral cross sections derived from the measured reaction rates. The utilization of the measured integral data for testing the respective cross sections on ENDF/B-V is covered in Section III. Both conventional integral testing based on a comparison of

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calculated-to-measured integral cross sections and testing based on least-squares-adjustment analyses are used to assess the consistency between the measured integral data and the evaluated cross sections. Conclusions drawn from this work are presented in Section IV.

II. Integral Data Base

The irradiation facility for the actinide integral measurements is the CFRMF[1,2] which is a zoned-core critical assembly with a fast neutron spectrum zone in the center of an enriched ^{235}U, water moderated thermal driver. Approximately 95% of the neutrons in the central spectrum are between 4 keV and 4 MeV and the median and mean neutron energies are 370 keV and 1760 keV, respectively. The central neutron spectrum is a Cross-Section Evaluation Working Group (CSEWG) benchmark field for testing dosimetry, fission-product and actinide cross sections for ENDF/B-V. An update of the CFRMF central neutron spectrum characterization has been reported recently[3,4].

A variety of techniques have been used for the integral measurements. Fission rates based on absolute fission chamber (FC) measurements have been reported by Grundl et. al[5] for ^{235}U, ^{238}U, ^{237}Np and ^{239}Pu. Measurements of the fission rates of ^{232}Th, ^{233}U and ^{240}Pu relative to the fission rate of ^{235}U have been reported by Gilliam and Rogers[6]. These latter experiments employed the NBS double fission chamber (DFC). The gamma spectrometric method[7] was used in the determination of the fission rates for ^{232}Th, ^{242}Pu, ^{241}Am and ^{243}Am based on absolute measurements of the gamma emission rates of the prominent lines in the ^{140}Ba-^{140}La decay[8,9,10]. With the exception of the capture rate for ^{241}Am which was determined using isotope-dilution-alpha spectrometry (IDAS)[10], the capture rates for ^{232}Th, ^{238}U, ^{242}Pu and ^{243}Am are based on gamma spectrometric measurements[7,10]. Many of the measurements were made as part of the Interlaboratory Reaction Rate (ILRR) Program[11].

Spectrum-averaged cross sections were derived by dividing the measured integral reaction rates by absolute neutron fluxes which were determined for each measurement. The neutron flux determinations are based on the use of the ^{197}Au(n,γ) ^{198}Au reaction as a power level monitor and on an independent determination of the neutron flux for one power level. The independent absolute flux value is based on a flux transfer[12] using measured fission rates for ^{239}Pu in the CFRMF and in the National Bureau of Standards (NBS) ^{252}Cf standard field and on a measured integral cross section for ^{239}Pu(n,f) in the ^{252}Cf field[7].

A tabulation of the "measured" integral cross sections is given in column 4 of Table 1. The value in parenthesis is the estimate of the total percent error in the integral cross section, at the one sigma confidence level. Columns 2 and 3 provide necessary identification of the experiment type and the reference for the integral reaction-rate measurements.

III. Cross-Section Data Testing

Integral tests of the evaluated capture and fission cross sections were made by comparing the "measured" integral cross sections to integral cross sections computed using 620-group representations of the CFRMF central spectrum[13] and of the differential data on ENDF/B-V. The calculated integral cross sections and the ratios of calculated-to-measured integral cross section are listed in columns 5 and 6, respectively, of Table 1. Two uncertainty values, expressed as percent, are listed in parenthesis for the calculated integral cross sections. The first corresponds to the error in the calculated integral cross section due to spectrum uncertainties[4] only. The second uncertainty contribution, wherever it is given, corresponds to the error in the calculated integral cross section due to cross-section uncertainties as processed from the ENDF/B-V uncertainty files. One uncertainty value, expressed as percent, is given for each C/M ratio. That value corresponds to the quadrature sum of the error in the "measured" integral cross section and the error in the calculated integral cross section due to spectrum error contributions only.

For the radiative capture reactions, the integral test analysis indicates an inconsistency between the measured integral data and the ENDF/B-V evaluated cross sections for ^{232}Th, ^{242}Pu, ^{241}Am and ^{243}Am. The integral test indicates that the ENDF/B-V capture cross section for ^{238}U is consistent with the measured integral data. Column 7 in Table 1 lists the ratio of the integral cross section computed with ENDF/B-V differential data to that computed with ENDF/B-IV differential data. This information indicates how the changes made to the capture cross section in going from ENDF/B-IV to ENDF/B-V impact the consistency test. The last two columns in Table 1 provide a qualitative indication of the response range for each reaction in the CFRMF spectrum. Response plots as a function of energy are given in Reference 14 for these reactions.

Least-squares-adjustment analyses were made with the FERRET code[15]. The adjustment analysis was made in a 53-group energy structure and it included the following input data: (1) 23 dosimeter integral reaction rates[3], (2) six integral reaction rates for capture and fission in ^{242}Pu, ^{241}Am, ^{243}Am, (3) CFRMF spectrum and associated covariance matrix, (4) cross sections processed with CFRMF spectrum weighting from ENDF/B-V for all reactions, and (4) covariance matrices for all reactions. The measured integral data were assumed to have zero correlation in the analyses. Covariance matrices for the dosimeter reactions were a mixture of matrices processed directly from the ENDF/B-V covariance files and matrices generated by F. Schmittroth to improve upon the ENDF/B-V prescriptions[16]. Covariance matrices for the actinide reactions were generated by using a gaussian-type parametric form to describe the short-range correlations between the group wise uncertainties as obtained from ENDF/B-V and adding an additional normalization uncertainty in the unresolved and smooth energy ranges. This approach was used because of the limitations of the error files on ENDF/B-V (no error files for ^{243}Am, missing error and correlation information for some energy regions for ^{241}Am and ^{242}Pu, block-type correlation specifications.) The ^{241}Am uncertainty information on ENDF/B-V was used for ^{243}Am, however, the normalization component was doubled from 15% for ^{241}Am to 30% for ^{243}Am.

Preliminary results of the least-squares-analysis are illustrated in Figures 1-3 for the ^{232}Th, ^{241}Am and ^{243}Am radiative capture cross sections. The upper part of each figure shows a direct comparison of the 53-group cross sections over the neutron energy ranges in which the CFRMF spectrum is sensitive. The bottom part of each figure shows the ratio of adjusted-to-unadjusted cross sections for the analysis. These figures indicate the energy range and the magnitude of the adjustments required to achieve consistency between the measured integral data and the evaluated cross sections. For $^{242}Pu(n,\gamma)$, the input covariance specification was too tight to permit sufficient cross-section adjustment to achieve consistency between the measured integral data and the adjusted cross section.

IV. Conclusions

In this paper we have summarized the actinide capture and fission integral data base for measurements in the fast neutron field of the CFRMF. An integral testing analysis in which the "measured" integral cross sections were compared to integral cross sections calculated with ENDF/B-V differential data indicated that for the fast neutron radiative capture cross sections for ^{232}Th, ^{242}Pu, ^{241}Am and ^{243}Am, the measured integral data are inconsistent with the evaluated cross sections. This same analysis indicated consistency between the measured integral data and the ENDF/B-V capture cross section for ^{238}U. A least-squares-adjustment analysis indicated that the following cross-section adjustments are required to resolve the discrepancies between the measured integral data and the ENDF/B-V capture evaluations: $^{232}Th(n,\gamma)$, ~ 5% to 10% up (1 keV to 17 MeV); $^{241}Am(n,\gamma)$, ~30% up (0.1 keV to 17 MeV), $^{243}Am(n,\gamma)$ ~ 44% up (0.1 keV to 17 MeV). This assessment for ^{232}Th is consistent with a recent measurement at 23-keV neutron energy by Baldwin and Knoll[17]. The above assessment for the ^{241}Am and ^{243}Am capture reactions is contingent on an experimental verification of the neutron-capture branching fraction data used in the analysis of the integral experiments[18]. However, the assessment of the ^{241}Am and ^{243}Am ENDF/B-V capture cross sections is consistent with other information presented at this meeting[19,20]. Although the present work indicated a significant discrepancy between the measured integral data and the ENDF/B-V capture cross section for ^{242}Pu, additional work is required to resolve questions concerning the accuracy of the measured integral data[15].

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Table 1. Summary of Integral Measurements and Integral Test Results for Actinide Reactions in the CFRMF

Reaction	Experiment Type	Reference	Integral Cross Section (mb) Measured	Calculated	C/M	V/IV	95% Response Range[a] E_L(MeV)	E_u(MeV)
^{232}Th(n,f)	γ-Spec	8	19.6(5.2)	19.14(7.5,5)	0.98(9)	1.06	1.2	8.3
(n,γ)	γ-Spec	8	290.8(3.8)	252.0(1.5,11)	0.87(4.1)	0.91	6.8 E-5	1.6
^{235}U(n,f)	FC	5	1538.(3.1)	1552.(0.5,2)	1.01(3.1)	0.99	2.2 E-4	3.6
^{238}U(n,f)	FC	5	75.1(3.3)	79.59(7.4,4)	1.06(8)	1.00	1.1	7.8
(n,γ)	γ-Spec	7	217.(3.7)	216.8(1.6,5)	1.00(4)	0.99	8.7 E-5	1.4
^{237}Np(n,f)	FC	5	548.(3.3)	606.1(3.6,10)	1.11(5)	1.01	0.3	5.8
^{239}Pu(n,f)	FC	5	1792.(2.2)	1773.(0.2,2)	0.99(2.2)	1.00	2.4 E-4	4.1
^{240}Pu(n,f)	DFC	6	573.(3.8)	623.4(6,-)	1.08(7)	1.01	9.0 E-2	5.6
^{242}Pu(n,f)	γ-Spec	8	557.(10)	477.1(6,-)	0.86(12)	0.96	0.3	5.9
(n,γ)	γ-Spec	8	146.(15)	266.1(2,-)	1.82(15)	1.18	5.2 E-5	1.3
^{241}Am(n,f)	γ-Spec	9	450.(6.2)	526.4(6,-)	1.17(9)	1.07	0.3	6.2
(n,γ)	IDAS	9	1550.(3.5)	1098.(2,-)	0.71(4)	1.69	1.2 E-4	1.0
^{243}Am(n,f)	γ-Spec	9	353.(6.1)	419.8(6,-)	1.19(9)	1.21	0.4	6.6
(n,γ)	γ-Spec	9	895.(4.8)	583.6(2,-)	0.65(5)	1.30	7.8 E-5	0.7

a. *Ninty-five percent of the reaction response in the CFRMF is between the lower energy, E_L, and the upper energy E_u.*

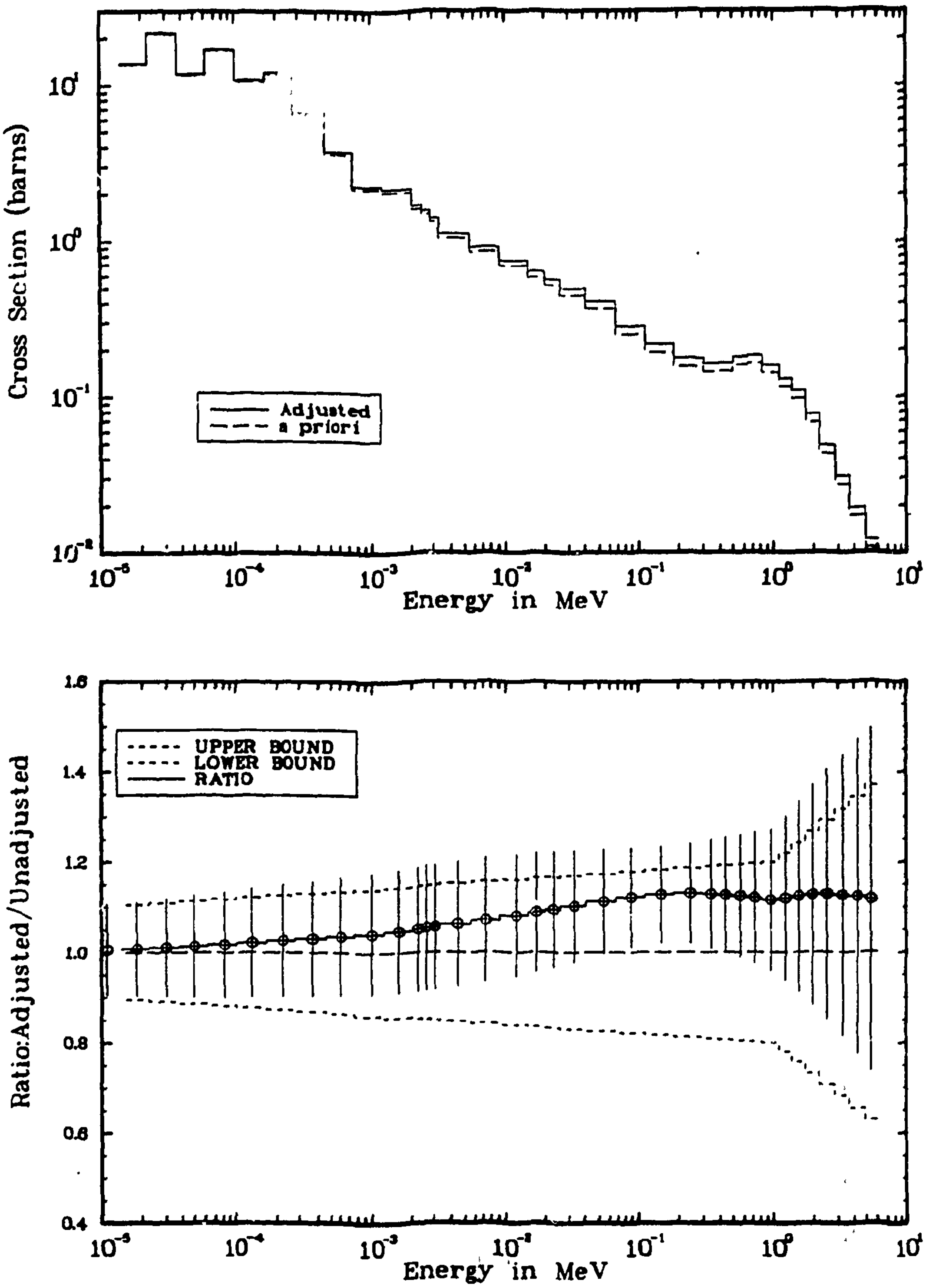

Figure 1. Comparison of adjusted and unadjusted cross sections for $^{232}Th(n,\gamma)$. In the lower half of the figure, input cross-section uncertainties are indicated by upper and lower bounds and adjusted uncertainties are indicated by vertical lines through the group mid-energy points.

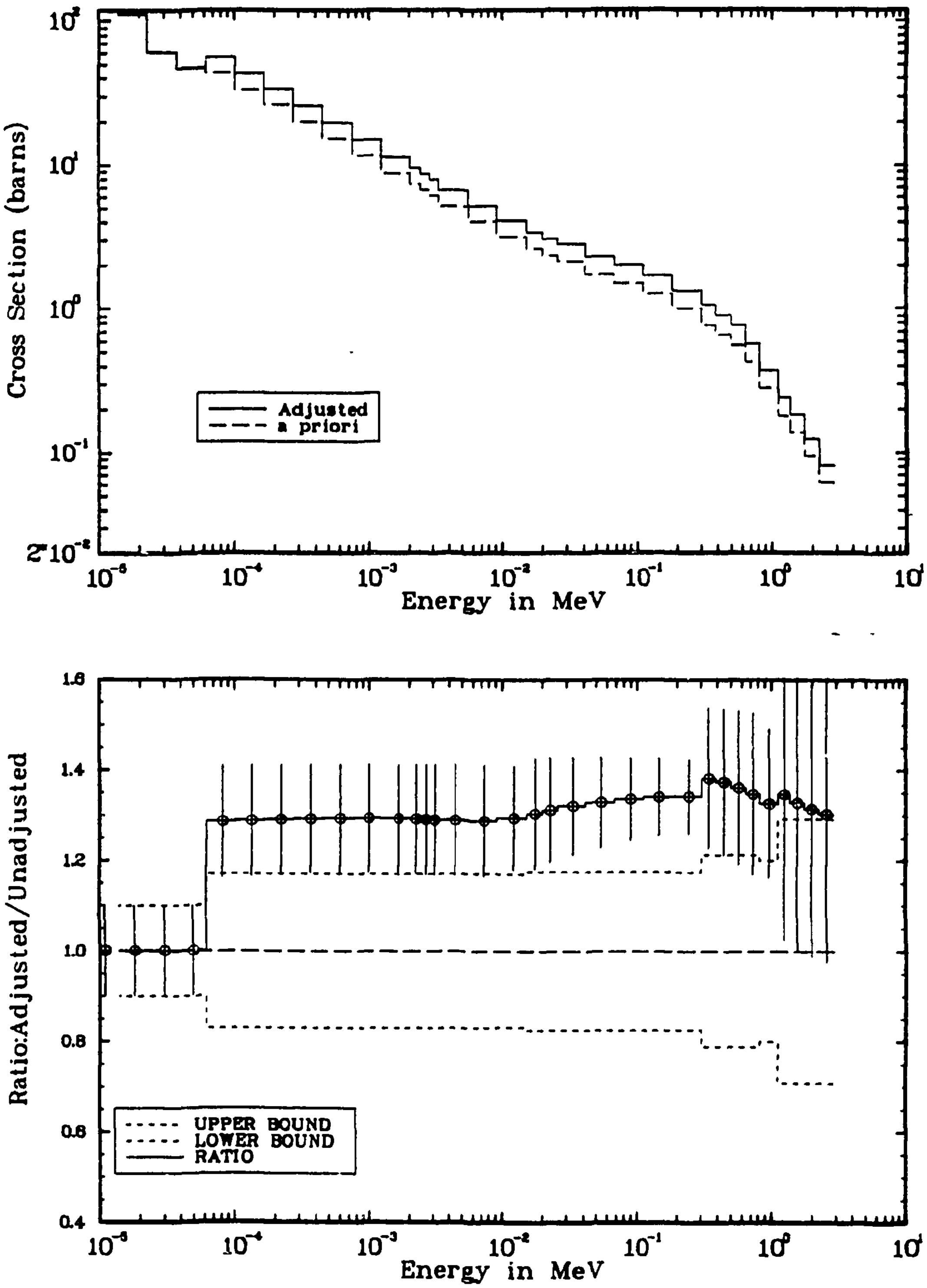

Figure 2. Comparison of adjusted and unadjusted cross sections for $^{241}Am(n,\gamma)$. In the lower half of the figure, input cross-section uncertainties are indicated by upper and lower bounds and adjusted uncertainties are indicated by vertical lines through the group mid-energy points.

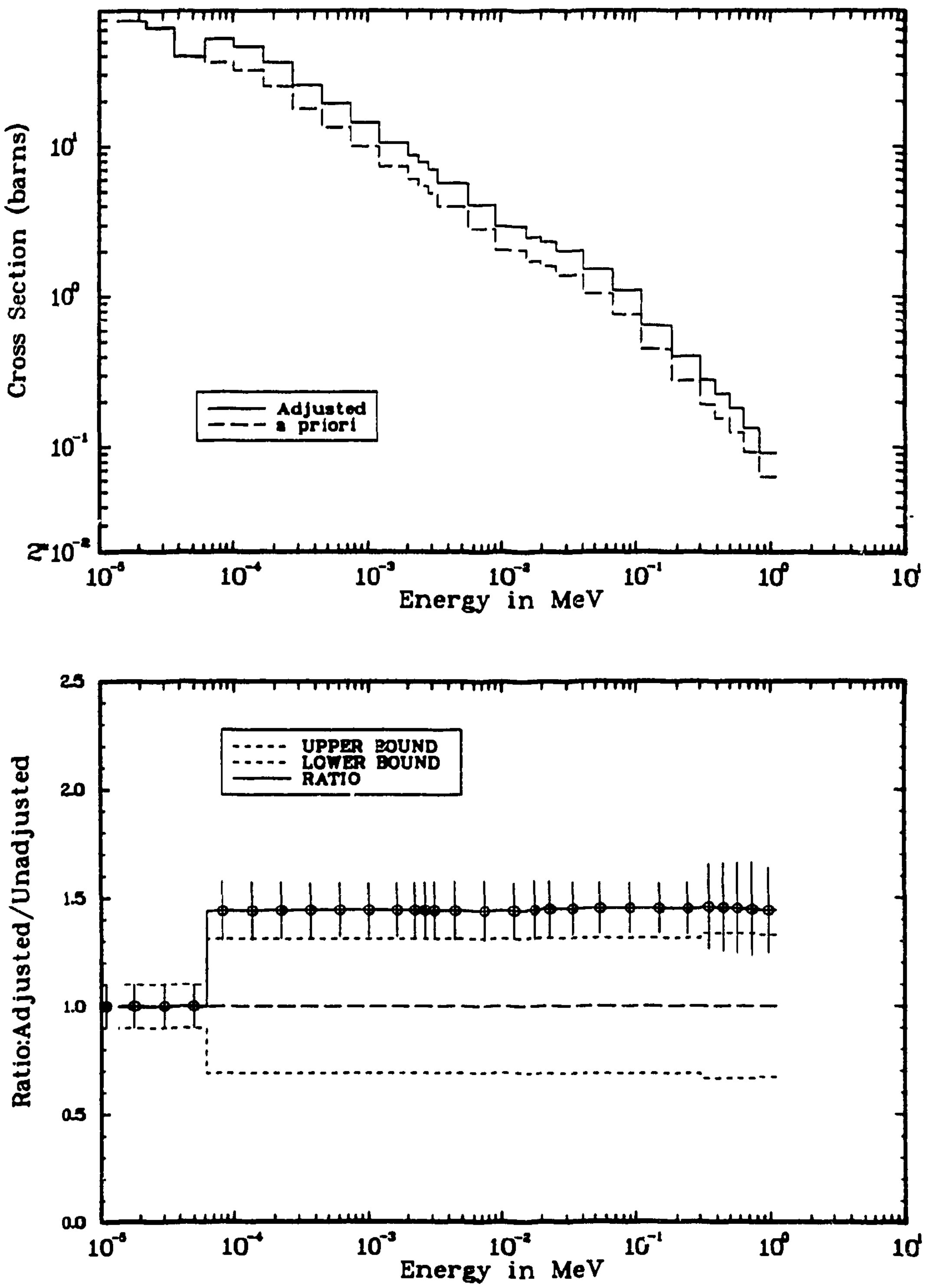

Figure 3. Comparison of adjusted and unadjusted cross sections for ^{243}Am(n,γ). In the lower half of the figure, input cross-section uncertainties are indicated by upper and lower bounds and adjusted uncertainties are indicated by vertical lines through the group mid-energy points.