Recent Cross-Section Evaluations in the Resonance Region at the Oak Ridge National Laboratory

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Abstract. The intent of this work is to present the results and to describe the procedures utilized to evaluate ^{233}U and ^{19}F for criticality safety applications. The evaluation was done in the resolved resonance region using the reduced Reich-Moore R-matrix formalism. The resonance analysis was performed with the multilevel R-matrix code SAMMY, which utilizes the generalized least-squares technique based on Bayes theory.

INTRODUCTION

In support of the U.S. Department of Energy (DOE) Nuclear Criticality Safety Program (NCSP), neutron resonance evaluations in the resolved resonance region were performed for ^{233}U in the neutron energy range up to 600 eV and for ^{19}F up to 1 MeV. Resonance parameters were derived by self-consistent analyses of the experimental data. The multilevel R-matrix code SAMMY[1] was used for the analysis of the data, including Doppler and resolution-broadening effects and other experimental effects. Compared with other fissile material, ^{233}U has a smaller critical mass. Criticality issues associated with the processing and disposal of ^{233}U require accurate knowledge of the ^{233}U cross-section data. Fluorine is an important nuclide that appears in several nuclear applications: for example, it is a component in the compound uranium hexafluoride used in the separation of the two isotopes of natural uranium, namely ^{235}U and ^{238}U. Fluorine is also an important material for molten salt coolants in reactor applications. Fluorine is an intermediate-mass nuclide that moderates neutrons in the epithermal energy range.

RESONANCE EVALUATION OF ^{233}U

The first multilevel-multichannel resonance analysis of the ^{233}U neutron cross sections was performed by Moore and Reich[2] and by Vogt[3] in the energy range below 12 eV. Bergen and Silbert[4] extended the analysis to the energy range of 20 to 60 eV. These analyses used both the single-level and multilevel-multichannel formalism; it was shown that the variation of the capture-to-fission ratio (α value) could not be reproduced by the single-level parameters. A more extensive work was performed by Reynolds and Steiglitz,[5] who used the least-squares fitting code MULTI[6] for the analysis of the fission and capture cross sections of Weston et al.[7] in the energy range thermal to 60 eV. The parameters of Reynolds and Steiglitz were converted to Adler-Adler parameters by deSaussure with the code POLLA[8] for the ENDF/B-V evaluated data file and are still used in the current version of the ENDF/B-VI. Single-level Breit-Wigner and Adler-Adler analyses were also performed by Kolar et al.,[9] Cao et al.,[10] and Nizamuddin.[11] The results of the single-level analysis of Nizamuddin et al. in the energy range from thermal to 100 eV were used with a large background contribution in the first version of ENDF/B-IV and JENDL-3. None of these evaluations proved to be satisfactory. The JAERI evaluation,[12] which is used in the current version of the Japanese Evaluated Nuclear Data Library (JENDL) and of the European file JEF, brought large improvements compared with the previous evaluations by allowing accurate calculation of the cross sections over the energy range up to 150 eV. The evaluation presented in this work extends the resonance energy range to 600 eV and improves the accuracy of the parameters by taking advantage of the excellent Oak Ridge National Laboratory (ORNL) experimental neutron transmission and fission data.

Experimental Data Base

To improve the accuracy of the ^{233}U resonance data at low energies and to extend the resolved resonance region up to 600 eV, new high-resolution transmission and fission experiments were performed at the Oak Ridge Linear Accelerator (ORELA). The neutron transmission measurements[13] were taken at the 79.8-m flight path with samples cooled to 11 K in order to reduce the width of the resonances by decreasing the Doppler broadening effect by a factor of 2 compared with the experiments conducted at room temperature; the ^{233}U sample was located 9 m from the neutron target. One series of measurements was done with a sample of 0.00298 at/b (with Cd filter in the beam) in the energy range 0.5 to 80 eV, and another series was performed with a sample of 0.0119 at/b (with ^{10}B filter in the beam) in the energy range 6 eV to 300 keV. The fission cross-section measurements[14] were performed at the 80-m flight path, consisting also of two series of measurements: one in the energy range 0.5 to 80 eV with a Cd filter and another in the energy range from 10 eV to 700 keV with a ^{10}B filter. Due to the flight path length and the excellent ORELA resolution, the experimental resolution was much better than any of the previous fission measurements. These new ORELA transmission and fission measurements were the primary data for the evaluation in the energy range 0.5 to 600 eV.

Most of the previous neutron cross-section measurements of ^{233}U in the thermal and resolved energy range were performed before 1975. The fission data were reviewed by Deruyter and Wagemans,[15] who found large discrepancies among the data, with differences of more than 50% on the average cross sections in some energy ranges and concluded that a consistent renormalization of all the data was not possible due to unknown important sources of systematic errors. In Ref. 12, the fission data of Deruyter and Wagemans, Weston et al.,[7] and Blons[16] were included in the experimental data base for the determination of the resonance parameters in the energy range up to 150 eV. It was shown that a renormalization of the three sets of data was possible with better than 3% accuracy, at least in the energy range analyzed, by taking as a reference the result of the most recent fission measurement by Wagemans et al.[17] normalized at thermal energy on the standard of Axton.[18] The experimental data of Weston et al. were the result of simultaneous measurement of the fission and the capture cross section; the Weston capture-cross section data are the only capture data available for the evaluation.

The experimental resolution parameters and the Doppler broadening parameters are needed for an accurate description of the shape of the experimental data by SAMMY. The parameters are found in the literature or obtained directly from the experimentalists who made the measurements. The accuracy of the resolution parameters was checked in the high-energy part of the data, where the width of the resolution function contributes to a significant portion of the observed width of the resonance or cluster of resonances. The effective temperature of the samples for the calculation of the Doppler broadening was taken to be 300 K, with an accuracy of about 10%.

For the data taken at liquid nitrogen temperature (Blons fission data) and at 11 K (ORELA neutron transmission data), the effective temperatures were those recommended by the experimentalists: i.e., 101 and 70.6 K, respectively. Since the evaluation included a large number of uncorrelated experimental data of different types (transmission, fission, and capture), it is unlikely that the errors in the temperature and resolution parameters could have a large effect on the accuracy of the resonance parameters. The most sensitive parameter is the capture width, for which one expects an accuracy of about 8% on the average value.

In addition to the microscopic or differential data [from time-of-flight (TOF) transmission or cross-section measurements], a variety of "integral quantities" may be fitted by SAMMY. These integral quantities are calculated by integrating over the microscopic absorption, fission, and capture cross sections and thus are a function of the resonance parameters. The derivatives of the integral quantities with respect to the resonance parameters are also calculated by SAMMY. The Maxwellian average cross sections at thermal energies are important for the interpretation of the thermal benchmarks. They are used in particular for the calculation of the Wescott g_x factors and the K1 parameter, defined, respectively, as

$$g_x = 2\,\sigma_x \Big/ \pi^{1/2}\,\sigma_{0x}$$

where g_x stands for fission (g_f) or absorption (g_a), and

$$K1 = \nu\sigma_{0f}\, g_f - \sigma_{0a}\, g_a$$

where σ_x and σ_{0x} are the Maxwellian-averaged cross sections and the cross sections at 0.0253 eV, respectively. Other important integral parameters are the fission and the capture resonance integrals, I_f and I_c, respectively. The resonance integral is defined as

$$I_x = \int_{0.5\,eV}^{20\,MeV} \sigma_x / E\; dE$$

Some evaluated ^{233}U integral data are given in Table 1.

TABLE 1. Evaluated integral quantities.

Quantity	ENDF/B-VI	Axton (Standard)	BNL	Present Work
g_a	0.9996 ± 0.0011	0.9995 ± 0.0011	0.9996 ± 0.0015	1.0033 ± 0.0011
g_f	0.9955 ± 0.0014	0.9955 ± 0.0014	0.9955 ± 0.0011	1.0004 ± 0.0014
I_a			897.0 ± 20.0	917.5 ± 20.0
I_f			760.0 ± 17.0	777.82 ± 17.0
K_1	742.0 ± 2.40	742.25 ± 2.37	738.0	746.77 ± 2.40
ν	2.4946 ± 0.0040	2.4950 ± 0.0040	2.493 ± 0.004	2.495 ± 0.004

The Results of the SAMMY Analysis

The resonance parameter file obtained from the SAMMY analysis of the experimental data in the energy range from thermal region to 600 eV contains 769 resonances, with 738 in the energy range analyzed and 31 external resonances. All of the resonances were considered as induced by s-wave neutrons. Small corrections should be applied to the calculated local s-wave strength function to take into account the effect of the unidentified small p-wave neutron resonances. The average spacing of the 738 resonances is 0.81 eV, which is much larger than the expected value. In the high-energy range of the data, it is no longer possible to determine resonance parameters for a single resonance. Instead, the resonance parameters represent multiplets of resonances and the absence of some resonances leads to larger average energy spacing.

The values of the cross sections calculated from the resonance parameters at 0.0253 eV are compared with the Axton standard and with ENDF/B-VI values in Table 2. The excellent agreement between Axton and the present calculation is due to the renormalization of the experimental fission and capture data to this standard prior to the SAMMY fit. A scattering-cross section file was also created in a small energy range near 0.0253 eV, according to the scattering cross section of Axton. The experimental total cross sections were not renormalized since they were the results of absolute measurements. The evaluated integral quantities shown in Table 1 and the values of thermal cross section indicated in Table 2 proved to be adequate for benchmark calculations in the thermal region.

TABLE 2. Thermal cross sections (barns) at 300 K.

	Present Results	Axton Standard	ENDF/B-VI
Fission	530.70 ± 1.34	530.70 ± 1.34	531.14 ± 1.33
Capture	45.22 ± 0.70	45.20 ± 0.70	45.51 ± 0.68
Scattering	12.18 ± 0.67	12.19 ± 0.67	12.13 ± 0.66

Comparisons of the experimental total and fission cross sections of Guber et al. with calculations using the resonance parameter derived in the ^{233}U evaluation are shown in Fig. 1 in the energy region 200 to 250 eV. The fit of the experimental data is very good.

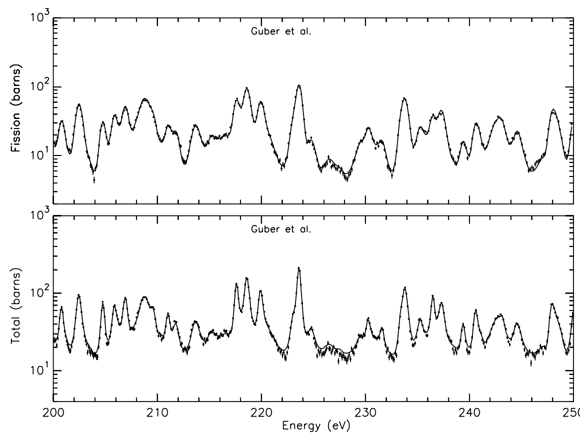

FIGURE 1. Comparison of the experimental total and fission cross sections of Guber et al. with calculations with SAMMY using ^{233}U resonance parameters.

RESONANCE EVALUATION OF ^{19}F

Resonance parameters for fluorine were determined by a self-consistent analysis of relevant experimental data, namely, high-resolution transmission data, capture cross-section data, and inelastic cross-section data. Thermal, total, and capture cross sections were also evaluated. The multilevel R-matrix code SAMMY[1] was used in the sequential analysis of the data. Two unique features of the ^{19}F analysis with SAMMY are the fitting of inelastic cross-section data using the Reich-Moore formalism and the analysis of the new ORELA capture cross-section measurements. In the energy region below 1 MeV, there are two inelastic energy levels at energies 109.9 and 197.2 keV, respectively. The Reich-Moore formalism was originally developed to accurately describe the fission cross section by taking into account the interference effect in the fission channels. Nevertheless, the Reich-

Moore formalism can be used to describe other reactions, such as the inelastic scattering cross section. In the analysis of the capture cross sections, multiple scattering effects were incorporated in the SAMMY evaluation.

Experimental Data Base

The experimental data used in the SAMMY evaluation of the ^{19}F cross sections are as follows:

(1) Three neutron transmission measurements of Larson et al.,[19] performed at the ORELA 80-m flight path, in the neutron energy range 5 eV to 20 MeV, with sample thicknesses of 0.13093 at/b (thick sample), 0.024184 at/b (medium sample), and 0.016886 at/b (thin sample). The thick and thin sample transmission data were analyzed in the energy region from 5 eV to 200 keV. The medium sample transmission data were analyzed in the energy region from 10 keV to 1 MeV. Both the medium and the thin samples contained carbon as well as fluorine; before the SAMMY analysis, the data were corrected for the carbon contribution. The total cross sections of carbon were parameterized according to the following formula,

$$\sigma = 4.743 - 3.175 \times E - 1.015 \times E^2 ,$$

where E is in MeV units. The carbon total cross section contribution was subtracted from the total effective cross section of ^{19}F corresponding to the medium and thin samples. Subsequently, a sequential SAMMY analysis of the experimental transmission data was performed by allowing a search on the normalization and constant background correction parameters. The normalization of the three transmissions was close to one. For the thin sample the background corrections was 0.00749; for the medium sample the background correction was 0.00013; for the thick sample the background was 0.00128. The effective cross section extracted from these transmission data was consistent with the existing ENDF/B evaluation.

(2) One capture cross-section measurement of Guber et al.[20] performed at the ORELA 40-m flight path in the energy range from 10 to 700 keV. The results of the normalization and background of the capture data calculated with the SAMMY code are, respectively, 0.99885 and 0.00009.

(3) Experimental data below 5 eV available in the literature were used for fitting the cross sections at low energy. The data of Rainwater et al.[21] were used to fit the total cross sections. The data span from 0.0205 to 37.1 eV. The only available capture data in the low-energy region are thermal energy data by Raman et al.[22] at the 8-MW Omega West reactor at Los Alamos, which were obtained by measuring the spectroscopy levels of ^{20}F.

(4) Inelastic cross-section measurements performed by Broder et al.,[23] at Obninsk (Russia), in the energy range up to 1 MeV. Three sets of inelastic cross-section measurements were included in the evaluation: (a) inelastic cross section corresponding to the first inelastic level at 109.9 keV with spin and parity 1/2-; (b) inelastic cross section corresponding to the second inelastic level at 197.2 keV with spin and parity 5/2+; and (c) total inelastic cross section, i.e., the sum of the cross section corresponding to the two inelastic levels.

SAMMY Analysis and Resonance Parameters

To initiate data fitting using the code SAMMY, two input files are needed in addition to the experimental data: namely, an input file containing initial estimates for values of the resonance parameters and an input file in which the information regarding spin groups is given. The initial set of Reich-Moore resonance parameters for ^{19}F up to 1 MeV was constructed as follows: (a) The resonance parameter values listed in the Brookhaven National Laboratory recommended data book[24] were used. The parameters consist of resonance energy E_r, neutron width Γ_n, gamma width Γ_γ, resonance spin J, and angular momentum l. (b) The peak search option of the RSAP code[25] was used to identify resonance energies and neutron widths from experimental total cross section data. (c) The SUGGEL computer code[26] was used to identify the orbital angular momentum l of the resonance levels. For the two inelastic channels, which open at the energies 109.9 and 197.2 keV, an arbitrary width value of 1000 meV was initially assigned. Natural fluorine is 100% ^{19}F, which has spin and parity $I^\pi = 1/2^+$.

The SAMMY analysis of the transmission, capture cross-section, and inelastic cross-section data from thermal to 1 MeV provided a set of resonance parameters with 2 s-wave, 5 p-wave, 17 d-wave, and 7 f-wave, for a total of 31 resonances. In addition to the 31 resonances, 4 external resonances were used: 1 negative-energy resonance to account for the bound levels, and 3 resonances above the cutoff energy of 1 MeV to account for the effect of the truncated levels above 1 MeV. Comparison of the experimental total and capture cross sections with the results of the SAMMY calculations using the resonance parameters is shown in Fig. 2.

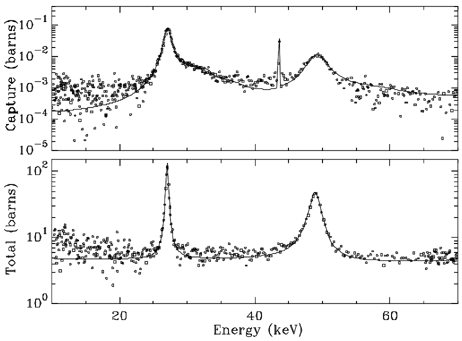

FIGURE 2. Comparison of the SAMMY calculations and experimental data for the capture cross section (top curve) and total cross section (bottom curve) in the energy region 40 to 70 keV.

Thermal Cross Section

The experimental data of Rainwater[21] were used for the fitting of the total cross section in the energy region from thermal to 5 eV. The Rainwater total cross section data were in agreement with the effective total cross section derived from the ORNL thick-sample transmission data. No data normalization was needed for the Rainwater total cross section. With the exception of the capture cross-section measurement of Raman et al.[22] at thermal energy, there are no other experimental capture cross-section data available in the literature. To fit the capture cross- section data at low energies, the ENDF/B-VI ^{19}F capture cross-section evaluation was used. The ENDF capture data were normalized at thermal to the value suggested by Raman. Figure 3 shows comparisons of the total and capture cross sections calculated with SAMMY using the resonance parameters with the experimental data. The top curve represents the fitting of the total cross-section data of Rainwater, and the bottom curve is the ENDF capture cross section normalized to the thermal value of Raman et al. The thermal cross-section values obtained with the resonance parameters derived in the evaluation of the ^{19}F are shown in Table 4. Also shown in Table 4 are the thermal cross-section values given by Mughabghab,[24] ENDF/B-VI release 8, JENDL, and the thermal capture cross-section value suggested by Raman et al.

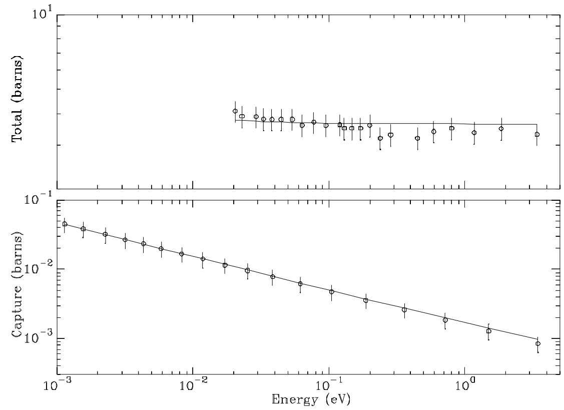

FIGURE 3. Comparisons of SAMMY theoretical calculations with the resonance parameters of the total cross section (top curve) and the capture cross section (bottom curve) below 5 eV with experimental data.

TABLE 4. Cross section for ^{19}F at 0.0253 eV.

	σ_t (barns)	σ_γ (barns)
Mughabghab	3.6506 ± 0.010	$(9.6 \pm 0.5) \times 10^{-3}$
ENDF/B-VI.8	3.750	9.500×10^{-3}
JENDL	3.511	9.570×10^{-3}
Raman	-	$(9.51 \pm 0.09) \times 10^{-3}$
ORNL	3.748 ± 0.004	$(9.51 \pm 0.05) \times 10^{-3}$

Conclusions

The Reich-Moore formalism of the SAMMY code was used in the evaluation of the neutron cross section of ^{233}U and ^{19}F. The evaluations include a large number of experimental data. Extension to a neutron energy range much larger than in previous evaluations was possible because of the recent TOF high-resolution neutron transmission, fission, and capture cross-section measurements performed at ORELA. The evaluations will be submitted for inclusion in the ENDF cross-section library.

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