Updated and revised neutron reaction data for ^{233}U

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Abstract: A complete set of n+^{233}U neutron reaction data from 10^{-5} eV–20 MeV is updated and revised based on the evaluated experimental data and the feedback information of various benchmark tests. The main revised quantities are nubars, cross sections as well as angular distributions, etc. The benchmark tests indicate that the present evaluated data achieve very promising results.

Key words: ^{233}U, nuclear data, evaluation, CENDL, neutron

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1 Introduction

For the improvement of nuclear energy, ^{233}U is an important material in Th/U fuel cycle applications. The neutron reaction data of ^{233}U play an important role in the framework of studies concerning new fuel cycles and nuclear waste disposal, as well as other nuclear fields [1–3].

A complete set of neutron reaction data of ^{233}U in CENDL-3.1 [4] was completed in 2002 and released in 2009. Simply reviewing the evaluated data of ^{233}U in CENDL-3.1 [4] can not satisfy the application requirements in some cases. The calculated effective neutron multiplication factor (k_{eff}) values with ^{233}U data in CENDL-3.1 [4] for critical spectra in spherical metal installations are around 0.2%–1.2% lower than the experimental ones. In light water breed reactors, the calculated k_{eff} values are different by ~0.5% from measurements. Therefore, the ^{233}U data in CENDL-3.1 [4] does not meet the higher accuracy requirements for nuclear science and engineering.

As many important measurements of cross sections for n+^{233}U were reported after 2002 and the international evaluation of neutron cross section standards [5] was updated and released in 2006, these factors must be taken into account in the modified evaluation of CENDL-3.1 [4]. In order to meet the requirements of new energy source development and nuclear science technology applications, the data of ^{233}U in CENDL-3.1 [4] should be reviewed and updated. A complete set of n+^{233}U neutron reaction data from 10^{-5} eV –20 MeV is successfully updated and revised, on the basis of the evaluated experimental data and the feedback information of various benchmark tests.

2 The resonance parameter and nubar

The complicated construction of cross sections in the resonance energy region is described using resonance parameters generally. The resolved resonance evaluation was performed in the energy range below 600 eV and based on the multi-level R-matrix analysis by L.C. Leal [6]. The unresolved resonance region evaluation covers the energy range from 600 eV to 40 keV. The above resonance parameters are adopted by this work. At 40 keV the total, (n, n), (n, f) and (n, γ) cross section values are adjusted slightly to join smoothly with the values calculated from the resonance parameters.

The total average neutron multiplicity per fission (total nubar) is a sum of average delayed (ν_{d}) and prompt (ν_{p}) neutron multiplicity per fission. The evaluated average neutron multiplicity per fission is based on the measurements of absolute ν and relative to ^{252}Cf neutron multiplicities from spontaneous fission. The available experimental data and systematics are used in nubar evaluation also. And the dependence of ν_{p} on incident neutron energy is given by an approximated linear function below 20 MeV. More detailed information on nubar evaluation is described in Ref. [9]. The evaluated results of ν_{d} and ν_{p} are compared with the measured and evaluated ones [7, 8, 10] as shown in Figs. 1 and 2. The results of the present evaluation agree well with the measurements.

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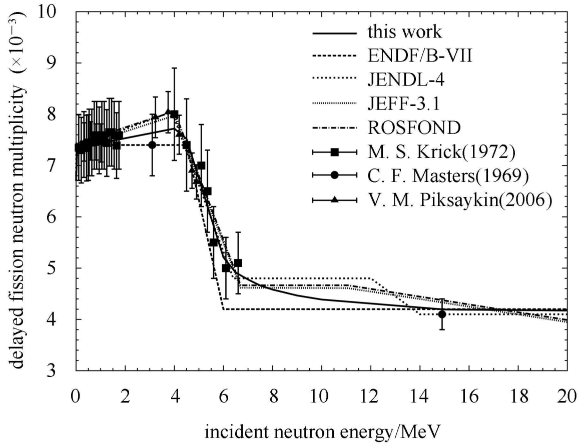

Fig. 1. The evaluated delayed fission neutron multiplicity, $\nu_{\rm d}$, compared with the measured and other evaluated data.

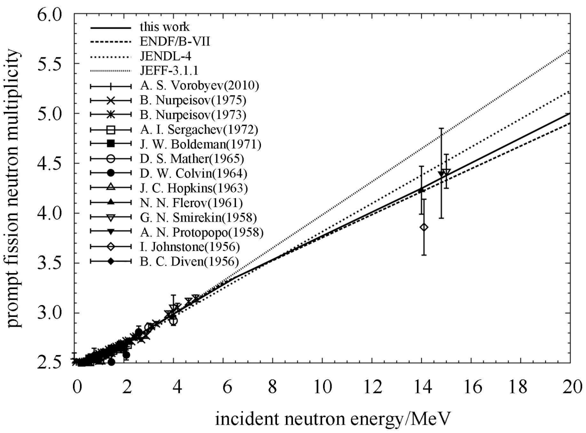

Fig. 2. The evaluated prompt fission neutron multiplicity, $\nu_{\rm p}$, compared with the measured and other evaluated data.

3 The reaction cross section above resonance energy region

3.1 The total cross section

There are a lot of experimental data up to 20 MeV. The data were measured by W.P. Poenitz [11, 12] from 0.058 to 4.43 MeV in 1978 and from 0.048 to 4.8 MeV in 1981 by using time of flight (TOF) technology with a 99.97% enriched ^{233}U sample. W.P. Poenitz [13] measured the total cross sections from 1.8 to 20 MeV in 1983, in which the energy region extends from 5 to 20 MeV comparing with their previous measurements. This evaluation is mainly based on the measurements of W. P. Poenitz [11–13], which are higher by 8% around 1.6 MeV than previous measured data. And the measured data of $\sigma_{\rm tot}$ cover the smooth energy regions from 40 keV to 20 MeV, which are compared with the evaluations as shown in Fig. 3.

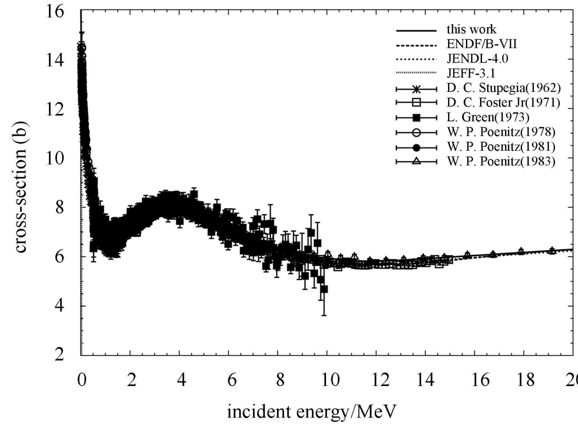

Fig. 3. Comparison of the evaluated data with the measurements for (n, tot).

3.2 The fission cross section

Lots of measurements for fission cross section exist using those relevant to "standard" or other accurately measured reactions. The discrepancy of measurements between different laboratories before 1975 is about 24%–28% around 6 MeV and missing measurements in some energy regions. In order to resolve the discrepancy of the measured data and fill the energy gaps, some accurate measurements were performed after 1975. The measured information of (n, f) is listed in Table 1.

J. W. Behrens [14] improved the experimental technology using the TOF technique with an ionization fission chamber in 1976. This method is the so-called "threshold cross section method". The advantage of this method [14] is that it does not require knowledge of the neutron flux and the efficiency of the fission chamber. Furthermore, this method is suitable for white neutron sources and accurate fission cross sections could be obtained in wide energy region using them. During 1976 $\sim$ 1978, the fission cross section ratios between ^{233}U and ^{235}U were measured by J. W. Behrens [14] in the energy region from 1.0 to 29 MeV and the ratios between ^{233}U and ^{238}U by G. W. Carlson [15] in the energy region from 0.85 keV to 29 MeV using the same method. Their data are consistent with each other within the uncertainty. This is a significant improvement for reducing either the experimental uncertainty or the discrepancy. The above measurements are re-normalized to the international evaluation of neutron cross section standards [5].

There are a lot of measurements [16–30] after 1978. The fission cross sections were provided in the measurement of B. Fursov [17] only, which is unable to make re-normalization due to a lack of information about the monitor. K. Kanda [18] used a back-to-back ionization chamber to measure the fission cross sections relative to ^{235}U, however he only estimated the lost counts by the bias setting and counting statistics for corrections.

Table 1. The measured information of fission cross sections.

year	author	E_n/MeV	sample	detector*	monitor	comments
1976	J. W. Behrens	1.–29.	enriched	FISCH	^{235}U(n,f)	TOF at linac
1977	V. M. Adamov	14.8		FISCH	Absolute	assop;more corrections
1978	G. W. Carlson	0.00085–29	enriched	FISCH	^{238}U(n,f)	TOF at linac
1978	W. P. Poenitz	0.137–8.1	99.47%	IOCH	Absolute	TOF
1978	B. I. Fursov	0.24–7.4	93.31%	IOCH	^{235}U(n,f)	absolute measure. CS ratio
1983	I. D. Alkhazov	14.7	metallic	FISCH	Absolute	assop; data from Fig.
1984	K. R. Zasadny	14.62		TRD	^{56}Fe(n,p)	corrected value
1986	K. Kanda	0.49–6.97	metallic	FISCH	^{235}U(n,f)	FISCH metallic sample
1988	F. Manabe	13.5–14.9		FISCH	^{235}U(n,f)	
1988	J. W. Meadows	14.74		IOCH	^{235}U(n,f)	TOF
1998	D. L. Shpak	0.02–6.38	93.31%	GLASD	^{235}U(n,f)	
1991	P. W. Lisowski	0.583–398		IOCH	^{235}U(n,f)	data from Fig; no information
2002	O. A. Shcherbakov	0.57–196.2		IOCH	^{235}U(n,f)	TOF; ^{235}U of JENDL-3.2
2004	F. Tovesson	1.0–7.5		FISCH	Absolute	^{237}Np(n,f) monitor for flux
2009	M. Calviani	3.4E-8–1.	99.01%	FISCH	^{235}U(n,f)	at n_TOF at CERN (185 m)

∗ FISCH: Multi-layer ionization chambers for fission fragments; IOCH: Ionization chamber; TRD: Polyester track detector; GLASD: Glass detector;

D. L. Shpak [19–21] measured the fission cross sections three times with the glass detector using p-^{7}Li, p-T neutron source at Van-de-Graaff accelerator during 1975–1998. The glass detector system was used and its average scanning error was around 0.4%. And various corrections also were done for neutron backgrounds from different sources. The results of three times measurements are coincide with each other very well, within 2%, and one or two oddity energy points were revised.

One high-accuracy fission cross-section measurement was performed by M. Calviani [25] using a white neutron source and n_TOF with an avalanche fission ionization chamber from ∼0.034 eV to 1 MeV neutron energy, in which the flight path is 200 m and then very precise energy discrimination is achieved. Enriched ^{233}U and ^{235}U samples were used and the corrections for different counting efficiencies originating from different thickness samples were made using the Monte Carlo simulation method. The dead-time correction was made also.

The fission cross section at 14.8 MeV was reported by V. M. Adamov [26] from CCPRI Lab using the associated particle method in 1977 and the collaborated result was published by I.D. Alkhazov [27] from GERDRE and CCPRI Lab at 14.7 MeV in 1983. However, the above two measurements have a big discrepancy, and correction and re-normalization were not performed due to a lack of information about the measurements. K. R. Zasadny [28] measured the fission cross section at 14.7 MeV in 1984 and detailed experimental information was given to enable correction and re-normalization of the results.

F. Manabe [29] measured the fission cross section ratios relative to ^{235}U around 14 MeV using a back-to-back parallel plate ionization chamber. The isotopic composition of the samples was determined by means of alpha spectroscopy. The various effects were analyzed and corrected in detail. J. W. Meadows [30] adopted a low mass double ionization chamber with ^{230,232}Th, ^{233,234,236,238}U, ^{237}Np and ^{239,242}Pu samples to measure the fission cross section ratios relative to ^{235}U at 14.8 MeV. Their measured results agree with other high accuracy measurements within uncertainty.

According to the above-mentioned experimental information, the present evaluation emphasizes the neutron energy region below 20 MeV. Therefore, the data measured by J. W. Behrens [14] and G. W. Carlson [15] with the “threshold cross section method” and W. P. Poenitz [16], D. L. Shpak [19–21], M. Calviani [25] and J. W. Meadows [30] are important data and add more weight. The measurement of J. W. Meadows [30] is regarded as the foundation around 14 MeV. The fission cross sections between 0.577 and 196 MeV were measured by O.A. Shcherbakov [23] using the TOF method with a fast parallel plate ionization chamber and a spallation neutron source. The measurements of P. W. Lisowski [22] and F. Tovesson [24] were spread around the accurate ones as the results belong to the process reports in 1991 and 2004, respectively. These measurements are used as a reference in evaluation also.

The present evaluated result is compared with the measurements and other evaluations shown in Fig. 4. The present evaluation agrees with ENDF/B-Ⅶ [7] and JENDL-4.0 [8], especially in the energy region below 10 MeV. There exists a small downhill strike from JENDL-4 [8] around 11.5 MeV.

3.3 The inelastic scattering cross section

The inelastic scattering cross section in fissile cases is much less accurate than the other reactions due to the lack of experimental data. The elastic scattering cross sections were measured by A.B. Smith [31] from 0.93 to 3.55 MeV. As a result, (n, n′) was deduced by subtracting (n, n), (n, f) and (n, γ) from (n, tot) and adjustment in CENDL-3.1 evaluation. Presently, reviewing the measurement of A.B. Smith [31], we find the elastic cross sections including the contribution of inelastic scattering from the partial low-lying excited states. Consequently, the elastic and inelastic scattering cross sections are overestimated and underestimated in CENDL-3.1 [4], respectively.

In this work, the elastic scattering cross sections are evaluated according to the high resolution measurement [32] as shown in Fig. 5. And the evaluation of (n, n′) reaction is based on the present evaluation of (n, n) and other dependent cross sections. The revised (n, n′) is compared with other evaluations as shown in Fig. 6. There is a very unusual shape below 1 MeV for ENDF/B-Ⅶ [7], which is very different from the present work and other evaluations.

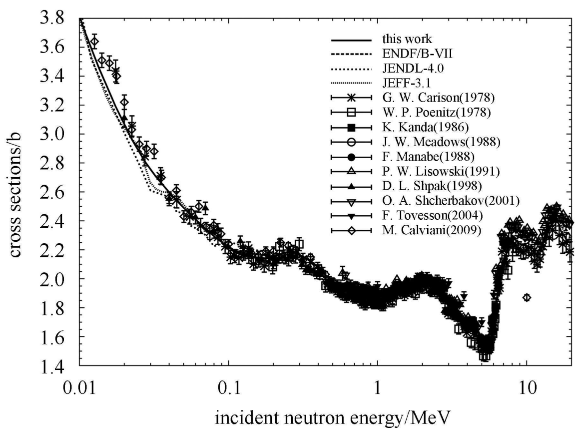

Fig. 4. Comparison of (n, f) reaction between the evaluations and measurements.

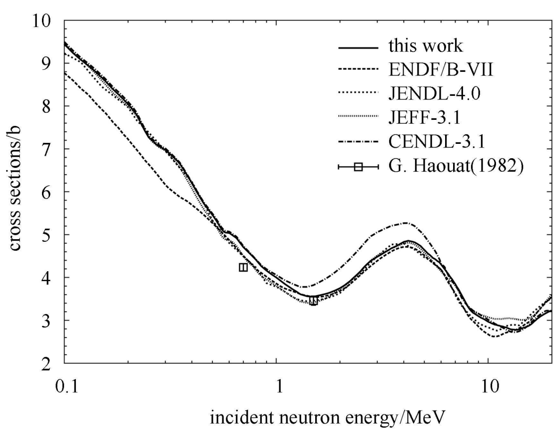

Fig. 5. Comparison of the evaluations for (n, n) reaction.

3.4 The radiation capture cross section

For a radiation capture reaction, some measurements are available from 40 keV to 1 MeV. The α values (=(n, γ)/(n, f)) were measured by J. C. Hopkins [33], and multiplied with fission cross sections to obtain (n, γ) data. Based on the theoretical calculation and experimental data, the evaluated data are obtained, which reproduce the experimental data very well as shown in Fig. 7.

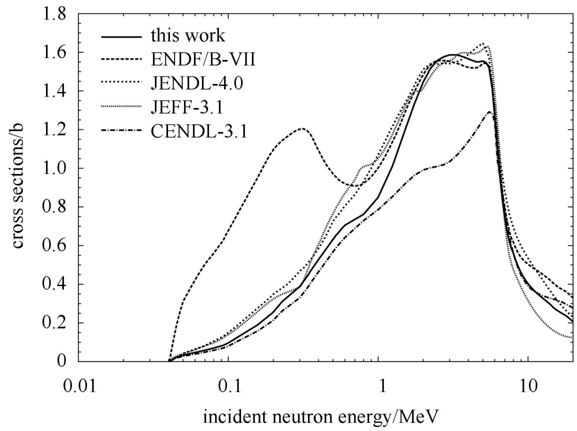

Fig. 6. Comparison of the evaluations for the (n, n′) reaction.

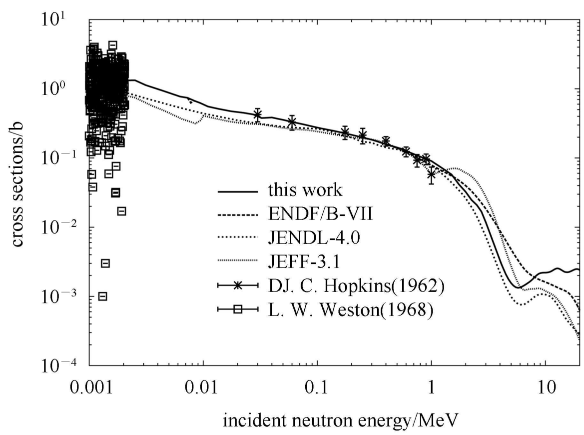

Fig. 7. Comparison of the evaluated and experimental data for (n, γ) reaction.

4 The theoretical calculation and data adjustment

There are only two sets of measurements for elastic scattering angular distribution at 0.7 and 1.5 MeV, which were carried out by G. Haouat [32]. As for nonelastic cross sections, the measurement was performed by R. F. Taschek [34] at 1.47 and 1.6 MeV. In order to obtain the optimal optical potential parameter, below the threshold energy of (n, 2n) reaction, the experimental data of the (n, γ)+(n, n′)+(n, f) reaction are used as the nonelastic cross sections. Finally, an optimum set of neutron optical potential parameters (OMP) for ^{233}U is obtained in the energy region 0.001 ~20 MeV. The theoretical calculation is performed with FUNF [35] code.

The Legendre coefficient for elastic scattering angular distribution is 22 orders starting at 10.25 MeV and 24 orders staring at 13.25 MeV, respectively. The calculated elastic scattering angular distribution reproduces the experimental data [32] well. The calculated result is compared with ENDF/B-Ⅶ [7], JENDL-4 [8] and JEFF-3.1 [10] at 18 MeV as shown in Fig. 8.

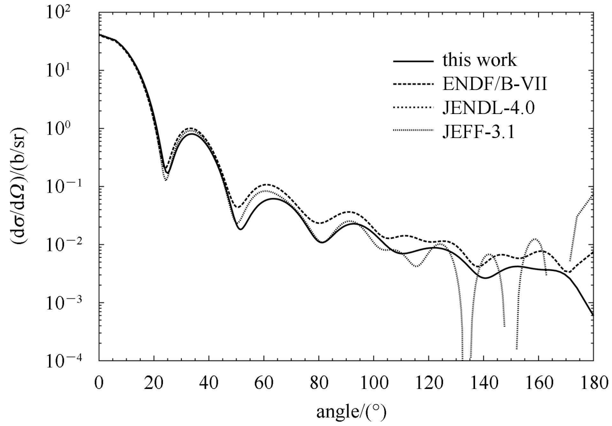

Fig. 8. Comparison of the elastic scattering angular distribution with other evaluations.

The inelastic scattering cross sections of ^{233}U in CENDL-3.1 [4] are updated and revised based on the re-analysis of the elastic scattering cross sections, and the new calculated result is obtained. To include the contribution of direct inelastic scattering, the 1st rotational band from the 1st to 3rd discrete levels are included in calculation using the ECIS [36] code. The adjusted parameters of level density, giant dipole resonance, fission barrier as well as an optimum set of OMP are used in the theoretical calculation. The reaction cross sections, secondary neutron spectra and angular distributions are obtained.

A complete set of neutron data is obtained based on the evaluated and calculated data. As an example, the calculated fission spectra at different incident neutron energies are shown in Fig. 9.

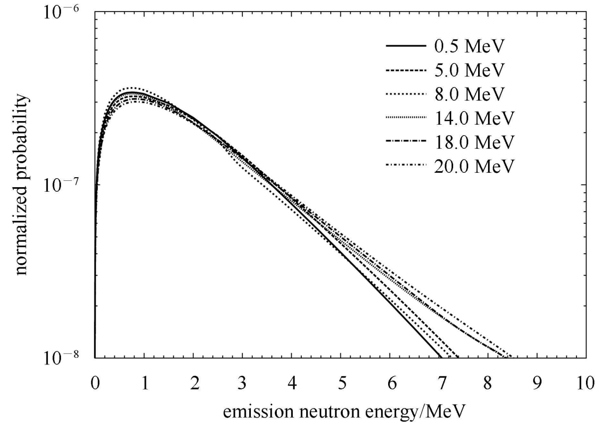

Fig. 9. The normalized secondary neutron spectra for fission reaction.

5 Summary

A complete set of n+^{233}U neutron reaction data from 10^{-5} eV–20 MeV is updated and revised. The present evaluation is compared with other evaluations [7, 8, 10] and measurements. From the point of view of the microscopic data, the present result is much better than the data in CENDL-3.1 [4], and reproduces the latest experimental data very well.

The promising results are obtained from feedback information of the predictions of benchmark when the data in CENDL-3.1 [4] are replaced by the present evaluated data. The calculated k_{eff} values are consistent with the measured ones of many benchmark testing installations within 0.1% in general. Presently, the evaluation data of ^{233}U could meet the requirements for nuclear science and engineering

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