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Re-evaluation of the $^{238}U(n,2n)^{237}U$ reaction excitation function from threshold to 30 MeV

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Cross section data for $^{238}U(n,2n)^{237}U$ reaction is needed for solving a wide spectrum of scientific and technical tasks.

The isotopic abundance of ^{238}U in natural uranium is (99.2742 ± 0.0010) atom percent [1], and the ^{237}U obtained via the (n,2n) reaction undergoes 100 % via β^- decay mode with a half-life of (6.75 ± 0.01) days. The $^{238}U(n,2n)^{237}U$ reaction rate is measured usually by detecting 59.5409-keV gamma radiation (I_γ = 0.345 ± 0.008) and 208.003-keV gamma radiation (I_γ = 0. 212 ± 0.003). Sometimes the $^{238}U(n,2n)^{237}U$ reaction rate is measured by detecting the 101.059-keV kα1 X-ray radiation ($I_{k\alpha1}$ = (0.245 ± 0.006). Recommended decay data for ^{237}U - half-life, X-ray and gamma emission probabilities per decay of ^{237}U were taken from Ref. [2].

In the process of evaluating cross sections and their uncertainties two common information sources were used for the $^{238}U(n,2n)^{237}U$ reaction: available differential and integral experimental data. Differential and integral experimental data were mainly taken from the EXFOR Library (Version, February 2014). In those cases where the needed information was absent in EXFOR, information was taken from the original publications.

Microscopic experimental data for the $^{238}U(n,2n)^{237}U$ reaction excitation function is given in the Refs [3-28] and cover the neutron energy range from 5.28 MeV to 19.0 MeV.

In the first step of evaluation all experimental data were analyzed. During this procedure, where possible, all experimental data were corrected to the new recommended cross section data for monitor reactions used in the measurements and to the new recommended decay data. The needed information about standards used for the correction of microscopic experimental data under investigation is given in the Table 1.

TABLE 1. DATA USED AS STANDARDS FOR CORRECTION OF MICROSCOPIC EXPERIMENTAL CROSS SECTIONS.

Monitor Reaction	**Cross sections used as standards**	**Half-life for residual nucleus**	**Radiation Mode and Energy**	**Emission Probability per decay**
$^{1}H(n,n)^{1}H$	Carlson+ [29]			
$^{27}Al(n,\alpha)^{24}Na$	Zolotarev [30]	14.9590 (12) H	Gamma 1368.63 keV	0.999936(15) [35]
$^{56}Fe(n,p)^{56}Mn$	Zolotarev [31]	2.5789 (1) H	Gamma 846.754 keV Gamma 1810.72 keV	0.9887 (3) [36] 0.2719 (79) [36]
$^{65}Cu(n,2n)^{64}Cu$	Zolotarev [32]	12.700 (2) H	Beta+ 653.1 keV Beta– 578.7 keV Gamma 511 keV Gamma 1345.77 keV	0.1740 (22) [37] 0.390 (4) [37] 0.348 (4) [37] 0.00473 (10) [37]
$^{93}Nb(n,2n)^{92m}Nb$	Zolotarev [33]	10.15 (2) D	Gamma 934.44 keV	0.9907 (4) [36]
$^{115}In(n,n')^{115m}In$	Zolotarev+ [34]	4.486 (4) H	Gamma 336.24 keV	0.458 (22) [38]
$^{169}Tm(n,2n)^{168}Tm$	Zolotarev [33]	93.1 (2) D	Gamma 198.25 keV	0.524 (16) [36]
$^{238}U(n,f)$	Carlson+ [29]			

For beta transition the end-point value of energy is given.

Experimental data of Refs. [3], [5-10], [13-28], in the process of this analysis were corrected to the new standards for the relevant monitor reactions (see Table 1) and to the recommended decay data for ^{237}U (Ref. [2]).

Experimental data of Kornilov et al. (Ref. [17]) were detailed in Refs [18] and [21]. Corrected to the new standards, the data of Refs [18] and [21] may be qualified as representative. The results of measurements in Refs [20], [22-23] may also be qualified as representative. The $^{238}U(n,2n)^{237}U$

reaction cross sections at 14.76 MeV in Ref. [20] were measured using two different monitor reactions ^{169}Tm(n,2n)^{168}Tm and ^{27}Al(n,a)^{24}Na. The obtained cross-section values of (731.19±40.14) mb and (716.44±30.95) mb agree well. Cross-sections measured by Konno et al. in the energy interval 13.31-14.86 MeV (Ref [22]) and by Filatenkov, Chuvaev in the energy interval 13.47-14.86 (Ref [23]) agree in the overlapping range in the limit of 1.2%.

Special correction has been applied to the experimental data of Refs [12] and [14-15]. Experimental data of Chou Youpu (Ref [12]) were renormalized to the cross-section value of 961.1 mb at 14.03 MeV, evaluated from the representative experimental data (Refs [20-23]). Frehaut et al. measured the ^{238}U(n,2n)^{237}U reaction cross section by using the D(d,n)He3 neutron source (Ref [14]) and the T(p,n)He3 neutron source (Ref. [15]). In addition to the correction due to the new standards, data of Frehaut et al. (Ref. [14]) were renormalized to the integral of the cross section that was calculated from experimental data by Filatenkov and Chuvaev (Ref. [23]) in the overlapping energy range 13.47 - 14.86 MeV, Fc = 1.09095. Experimental data by of Frehaut et al. (Ref. [15]) obtained in measurements with the T(p,n)He3 source in the energy range 6.49 - 13.09 MeV have been multiplied for the same factor.

The data base used for evaluation of the ^{238}U(n,2n)^{237}U reaction excitation function in the energy range from threshold to 30 MeV was formed from the experimental cross section data of Refs [5-7], [9-16], [18-23], [27-28]. The relative shape of the excitation function above 19 MeV was taken from the TENDL-2011 library [39]. The absolute cross sections used as input data in the interval 19-30 MeV were calculated from TENDL-2011 data by normalizing at 19 MeV to a value of 273.0 mb measured by Veeser and Arthur (Ref. [11]).

Experimental data obtained at TUNL (see table 2) were received from authors at the beginning of March 2015 and have, unfortunately, not been taken into account in the re-evaluation the ^{238}U(n,2n)^{237}U reaction excitation function. Data measured at the neutron energies 10.41, 10.92, 11.42, 11.42 and 14.80 MeV are needed in the analysis of decay parameters used for monitor reaction ^{197}Au(n,2n)^{196}Au.

TABLE 2. ORIGINAL AND CORRECTED EXPERIMENTAL DATA OF THE ^{238}U(N,2N)^{237}U REACTION MEASURED BY KRISHICHAYAN, M.BHIKE, W.TORNOW AT TUNL USING THE ACTIVATION METHOD. PRIVATE COMMUNICATION, MARCH 2015

E_n, MeV (mean)	Monitor cross-section, mb				^{238}U(n,2n)^{237}U cross-section, mb	
	^{27}Al(n,α)^{24}Na Original	IRDFF	^{197}Au(n,2n)^{196}Au Original	IRDFF	Original	Corrected
6.34	5.2	3.90			68.0 ± 3.00	51.00 ± 2.48
6.89	15.1	13.90			386.0 ± 12.0	355. 32 ± 12.54
7.40	26.0	25.14			824.0 ± 24.0	796.74 ± 25.02
7.87	38.1	36.58			1140.0 ± 34.0	1094.52 ± 34.15
8.38	52.0	50.85			1270.0 ± 37.0	1241.91 ± 37.63
8.89	66.0	65.32			1360.0 ± 36.0	1345.99 ± 37.55
9.40	78.5	78.28			1370.0 ± 35.0	1366.16 ± 36.89
9.91	89.0	89.05	969.4	977.78	1450.0 ± 38.0	1450.81 ± 40.19

$E_{n,}$ MeV (mean)	Monitor cross-section, mb			$^{238}U(n,2n)^{237}U$ cross-section, mb	
	$^{27}Al(n,\alpha)^{24}Na$ Original IRDFF	$^{197}Au(n,2n)^{196}Au$ Original	IRDFF	Original	Corrected
10.41		1217.8	1226.11	1420.0 ± 37.0	1429.69 ± 56.90
10.92		1416.4	1426.52	1440.0 ± 37.0	1450.29 ± 55.11
11.42		1575.7	1583.87	1440.0 ± 38.0	1447.47 ± 54.57
11.93		1710.9	1720.28	1430.0 ± 39.0	1437.84 ± 54.06
14.80		2162.23	2164.53	660.0 ± 20.0	660.70 ± 21.14

IRDFF – IRDFF-v1.05 data file.
Experimental data were corrected for monitor cross-sections only.
The $^{238}U(n,2n)^{237}U$ cross section at 9.91 MeV is given as measured relative $^{27}Al(n,\alpha)^{24}Na$ monitor.
Uncertainty in the corrected TUNL data takes into account uncertainty in the monitor cross section.

Experimental data of Refs [1], [3], [6] and Mather et al. at 12.4 MeV (Ref. [9]) were rejected due to inconsistency with the main bulk of experimental data. Experimental data given in the resent publications (Refs [22-24]) were also rejected due to the systematic underestimation of the $^{238}U(n,2n)^{237}U$ reaction cross section in the 14-MeV region.

Evaluation of the $^{238}U(n,2n)^{237}U$ reaction excitation function from threshold to 30 MeV was carried out by means of the generalized least-squares method within the PADE-2 code (Ref. [40]). The rational function was used as the model function (Ref. [41]). The method of statistical analysis of correlated data used to evaluate the excitation function of the $^{238}U(n,2n)^{237}U$ reaction was described in IAEA Reports of Refs [30], [32-33].

Uncertainties in cross sections for the evaluated $^{238}U(n,2n)^{237}U$ reaction excitation function are given in the form of a relative covariance matrix for 49-neutron energy groups (LB = 5). The covariance matrix of uncertainties was calculated simultaneously with the recommended cross-section data by means of the PADE-2 code and tested additionally by means of the COVEIG code (Ref. [42]). Six-digit eigenvalues of the relative covariance matrix in File-33 are as follows:

```
3.54537E-07     3.56403E-07     3.60136E-07     3.64977E-07
3.70250E-07     3.76599E-07     3.83985E-07     3.92169E-07
4.01013E-07     4.10394E-07     4.20574E-07     4.32294E-07
4.46724E-07     4.64905E-07     4.88137E-07     5.16198E-07
5.47091E-07     5.91668E-07     6.64585E-07     7.58874E-07
8.48397E-07     1.06058E-06     1.25714E-06     1.65012E-06
1.99413E-06     2.82910E-06     3.48385E-06     4.21256E-06
5.47387E-06     6.86460E-06     8.28105E-06     9.67752E-06
1.10246E-05     1.23039E-05     1.34765E-05     1.55533E-05
2.02276E-05     2.51922E-05     3.04420E-05     8.62659E-04
1.29487E-03     1.79071E-03     1.89912E-03     2.62241E-03
2.86479E-03     8.38884E-03     3.20631E-02     8.31231E-02
1.08298E-01
```

Evaluated group cross sections and their uncertainties for the excitation function of the $^{238}U(n,2n)^{237}U$ reaction are listed in Table 3. Group boundaries are the same as in File-33.

TABLE 3. EVALUATED CROSS SECTIONS AND THEIR UNCERTAINTIES FOR THE $^{238}U(n,2n)^{237}U$ REACTION IN THE NEUTRON ENERGY RANGE FROM THRESHOLD TO 90 MeV.

Neutron energy (MeV) from to	Cross section (mb)	Uncer-tainty (%)	Neutron energy (MeV) from to	Cross section (mb)	Uncer-tainty (%)
6.180 - 6.400	6.522	31.57	11.250 - 11.500	1529.000	2.44
6.400 - 6.600	59.639	6.78	11.500 - 11.750	1531.300	2.55
6.600 - 6.800	191.855	3.85	11.750 - 12.000	1524.650	2.66
6.800 - 7.000	361.291	3.06	12.000 - 12.500	1490.710	2.73
7.000 - 7.200	537.044	2.72	12.500 - 13.000	1397.690	2.63
7.200 - 7.400	700.581	2.61	13.000 - 13.500	1250.310	2.28
7.400 - 7.600	843.228	2.45	13.500 - 14.000	1071.220	1.91
7.600 - 7.800	962.739	2.27	14.000 - 14.500	891.820	1.75
7.800 - 8.000	1060.410	2.15	14.500 - 15.000	735.366	2.04
8.000 - 8.200	1139.100	2.10	15.000 - 15.500	610.597	2.85
8.200 - 8.400	1202.110	2.10	15.500 - 16.000	515.670	3.93
8.400 - 8.600	1252.590	2.10	16.000 - 16.500	444.987	5.00
8.600 - 8.800	1293.320	2.09	16.500 - 17.000	392.462	5.90
8.800 - 9.000	1326.610	2.06	17.000 - 17.500	353.057	6.60
9.000 - 9.200	1354.340	2.02	17.500 - 18.000	323.008	7.11
9.200 - 9.400	1378.000	1.98	18.000 - 18.500	299.632	7.48
9.400 - 9.600	1398.770	1.96	18.500 - 19.000	281.051	7.77
9.600 - 9.800	1417.510	1.95	19.000 - 19.500	265.955	8.04
9.800 - 10.000	1434.870	1.96	19.500 - 20.000	253.430	8.30
10.000 - 10.200	1451.250	1.99	20.000 - 22.000	230.285	9.05
10.200 - 10.400	1466.870	2.03	22.000 - 24.000	203.953	10.70
10.400 - 10.600	1481.730	2.09	24.000 - 26.000	185.278	12.46
10.600 - 10.800	1495.680	2.16	26.000 - 28.000	170.602	14.08
10.800 - 11.000	1508.310	2.23	28.000 - 30.000	158.431	15.51
11.000 - 11.250	1520.130	2.32			

Uncertainties in the evaluated $^{238}U(n,2n)^{237}U$ excitation function range from 1.75% to 31.57%. The highest uncertainty equal to 31.57% characterize data in the interval 6.18 – 6.4 MeV. The smallest uncertainties in the evaluated cross sections 1.75-1.91% are observed in the neutron energy range from 13.5 to 14.5 MeV. Uncertainties in cross sections exceeding 10% are in the energy range 22 - 30 MeV.

The evaluated excitation function for the $^{238}U(n,2n)^{237}U$ reaction in the neutron energy range from threshold to 30 MeV is shown in Fig. 1 in comparison with the equivalent data from the ENDF/B-VII.1 library and corrected experimental data. The same information, but in the narrow neutron energy ranges from threshold to 12 MeV and 12 to 16 MeV is shown in Fig. 2 and Fig. 3, respectively.

It should be noted that new TUNL measurements which were carried out relative to the $^{27}Al(n,\alpha)^{24}Na$ monitor reaction agree with the re-evaluated the $^{238}U(n,2n)^{237}U$ reaction excitation function in the energy range 6.89 – 9.91 MeV.

Integral experimental data for the $^{238}U(n,2n)^{237}U$ reaction are given in Refs. [43], [44], [45], [46]. Integral cross sections presented in Refs [43], [44] were measured in a neutron fields with similar spectra to the ^{235}U thermal fission neutron spectrum. Experiments which had been performed in a

^{252}Cf spontaneous fission neutron spectrum are described in Refs [45] and [46]. All of the experimental data were obtained by means of the activation method.
Experimental data of works [43], [44], [45] and [46] in the process of analysis were corrected to the new standards for the relevant monitor reactions (see Table 4) and to the recommended decay data for ^{237}U [2].

Recommended cross section data for monitor reactions used in the measurements of integral cross sections in ^{235}U thermal fission and ^{252}Cf spontaneous fission neutron spectra were taken from Refs. [47], [48], [49]. Digital data for ^{235}U thermal fission and ^{252}Cf spontaneous fission neutron spectra were taken from Refs. [50] and [51], respectively.

TABLE 4. DATA USED AS STANDARDS FOR CORRECTION OF INTEGRAL EXPERIMENTAL CROSS SECTIONS MEASURED IN ^{235}U THERMAL FISSION AND ^{252}CF SPONTANEOUS FISSION NEUTRON SPECTRA.

Monitor Reaction	Cross section used as standard, mb	Half-life for residual nucleus	Radiation Mode and Energy	Emission Probability per decay
^{27}Al(n,α)^{24}Na	0.7007±1.28% [47] U 1.016±1.28% [48] Cf	14.997 (12)H	Gamma 1368.63 keV	0.999936(15) [35]
^{46}Ti(n,p)^{46}Sc	11.51±1.70% [47] U	9.458 (12) M	Gamma 843.76 keV Gamma 1014.44 keV	0.718 (4) [36] 0.280 (4) [36]
^{54}Fe(n,p)^{54}Mn	78.09±1.50% [49] U	2.5789 (1) H	Gamma 846.754 keV Gamma 1810.72 keV	0.9887 (3) [36] 0.2719 (79) [36]
^{58}Ni(n,p)^{58}Co	108.2±1.30% [47] U	78.82 (3) D	Gamma 511 keV Gamma 810.78 keV	0.298 (4) [36] 0.99448 (8) [36]
^{115}In(n,n’)^{115m}In	187.8±1.23% [47] U	4.486 (4) H	Gamma 336.24 keV	0.458 (22) [38]
^{238}U(n,f)	325.7±1.64% [48] Cf			

Symbol “U” – means ^{235}U thermal fission neutron spectrum,
Symbol “Cf” – means ^{252}Cf spontaneous fission neutron spectrum.

Kobayashi et al. (Ref. [43]) measured the integral cross section at the fast reactor YAYOI of RRI at Kyoto University. Measurements were carried out in the core centre of the reactor. The neutron flux was monitored by the three dosimetry reactions ^{27}Al(n,α)^{24}Na, ^{58}Ni(n,p)^{58}Co and ^{115}In(n,n')^{115m}In. The integral cross section value of the ^{238}U(n,2n)^{237}U reaction- $\langle\sigma\rangle_{U\text{-}235}$ = (15.70 ± 0.80) mb measured by Kobayashi et al. was renormalized to the new standards. The second amendment was introduced to the new recommended value of the intensity of the 208.003-keV gamma radiation ^{237}U - I_γ = (45.8 ± 2.2)% [2], $F_{i\gamma}$ = (45.8/0.50) = 0.916. After correction to the new standards the integral cross section obtained in this experiment is equal to $\langle\sigma\rangle_{U\text{-}235}$ = (18.16 ± 0.93) mb.

Hashimoto et al. (Ref. [44]) measured the integral cross section of ^{238}U(n,2n)^{237}U reaction in the core of the KUR reactor. A slightly depleted uranium in the form U_3O_8 was used as a target for neutron irradiation. The uranium oxide, about 1 mg, was sealed into a quartz ampoule and irradiated along with flux monitor samples for 48 hours in the hydraulic conveyor tube in the KUR-core at 5000 kW reactor power. The neutron flux was determined by three dosimetry reactions, ^{46}Ti(n,p)^{46}Sc, ^{54}Fe(n,p)^{54}Mn and ^{58}Ni(n,p)^{58}Co. After irradiation and twenty days of cooling the chemical purification of the uranium sample was performed. The purified uranium was electrodeposited on a stainless steel plate. The absolute amount of ^{237}U, produced by the ^{238}U(n,2n)^{237}U reaction, was determined via registration of 208 keV gamma radiation. The integral cross section value of the ^{238}U(n,2n)^{237}U reaction- $\langle\sigma\rangle_{U\text{-}235}$ = (16.90 ± 1.40) mb measured by

Hashimoto et al. was renormalized to the new standards. After correction to the new standards, the integral cross section obtained in this experiment is equal to $<\sigma>_{U-235}$ = (18. 98 ± 1.57) mb.

The 300 micro-g Cf-252 source was used by Shani for measurement of the integral cross section of the $^{238}U(n,2n)^{237}U$ reaction (Ref. [45]). The half-inch (12.7 mm) diameter depleted (0.3% of ^{235}U) U foils were irradiated with the Al monitor foils for 7 days. The $^{238}U(n,2n)^{237}U$ reaction rate was determined via registration of 208 keV gamma radiation by the Ge(Li) detector. The background subtraction was made with an identical non-activated foil. The monitor $^{27}Al(n,\alpha)^{24}Na$ reaction rate was determined via registration of 1368.6 keV gamma radiation. After correction to the new standards, the integral cross section obtained in this experiment is equal to $<\sigma>_{Cf-252}$ = (18. 55 ± 2.28) mb.

Blinov et al. (Ref. [46]) from the Khlopin Radium Institute determined the integral cross section of the $^{238}U(n,2n)^{237}U$ reaction in the Cf-252 spontaneous fission neutron spectrum in two runs of measurements. The U samples were prepared from U3-O8 with ^{238}U content of 99.999 %. The weight of powder in each sample was equal to 2.5 grams. The first series of measurements has been carried out with two Cf-252 sources with a total weight of 18 micro grams and $^{238}U(n,f)$ reaction as flux monitor. The second series of measurements has been carried out with tree Cf-252 sources with a total weight of 50 micro grams and the $^{27}Al(n,\alpha)^{24}Na$ reaction as flux monitor. The $^{238}U(n,2n)^{237}U$ and $^{27}Al(n,\alpha)^{24}Na$ reaction rates were determined via registration of 208 keV and 1368.63 keV gamma rays by the Ge(Li) detector. The U-238(n,f) reaction rate was measured by a fission chamber. Two methods have been used to reduce the background under the 208 keV gamma-radiation of U-237: an increase of the cooling time and radiochemical purification of the irradiated samples from fragments and transuranium elements. The integral cross section value of the $^{238}U(n,2n)^{237}U$ reaction- $<\sigma>_{Cf-252}$ = (19.20 ± 1.90) mb measured by Blinov et al. was renormalized to the new standards. After correction to the new standards, the integral cross section obtained in this experiment is equal to $<\sigma>_{Cf-252}$ = (20.61 ± 2.02) mb.

The evaluated excitation function for the $^{238}U(n,2n)^{237}U$ reaction was tested against the above mentioned integral experimental data (Refs [43-44] and [46]). The calculated averaged cross sections over ^{235}U thermal fission neutron spectrum and ^{252}Cf spontaneous fission neutron spectrum are compared with the ENDF/B-VII.1 and experimental data in Table 5. The value of C/E given in the last column is the ratio of the calculated to experimental cross sections.

TABLE 5. CALCULATED AND MEASURED AVERAGED CROSS SECTIONS OF THE $^{238}U(N,2N)^{237}U$ REACTION IN ^{235}U THERMAL FISSION AND ^{252}CF SPONTANEOUS FISSION NEUTRON SPECTRA.

Type of neutron field	Averaged cross section, mb		90% responce function, MeV	C/E
	Calculated	Measured		
^{235}U thermal fission neutron spectrum	14.708 [A]	18.16 ± 0.93 [43]	6.80 – 11.1	0.80991
	15.466 [B]	18.98 ± 1.57 [44]	6.80 – 11.2	0. 85165
^{252}Cf spontaneous fission neutron spectrum	20.607 [A]	20.61 ± 2.02 [46]	6.80 – 11.5	0.99985
	21.363 [B]		6.80 – 11.6	1.03654

[A] - new re-evaluation
[B] - ENDF/B-VII.1

The C/E values obtained for the ^{252}Cf spontaneous fission neutron spectrum show that the $^{238}U(n,2n)^{239}U$ integral cross sections calculated from the newly evaluated excitation function agrees well with experimental data of Blinov et al. (Ref. [46]). The integral cross section calculated from the ENDF/B-VII.1 excitation function is a 3.6% higher than experimental value.

The C/E values obtained for the ^{235}U thermal fission neutron spectrum show a very big discrepancy between experimental and calculated data. This may be explained by the assumption that the measured cross sections $<\sigma>_{U-235}$ in Refs [43] and [44] are the total values of the two reactions $^{238}U(n,2n)^{237}U$ and $^{238}U(\gamma,n)^{237}U$, where the latter was present as an impurity.

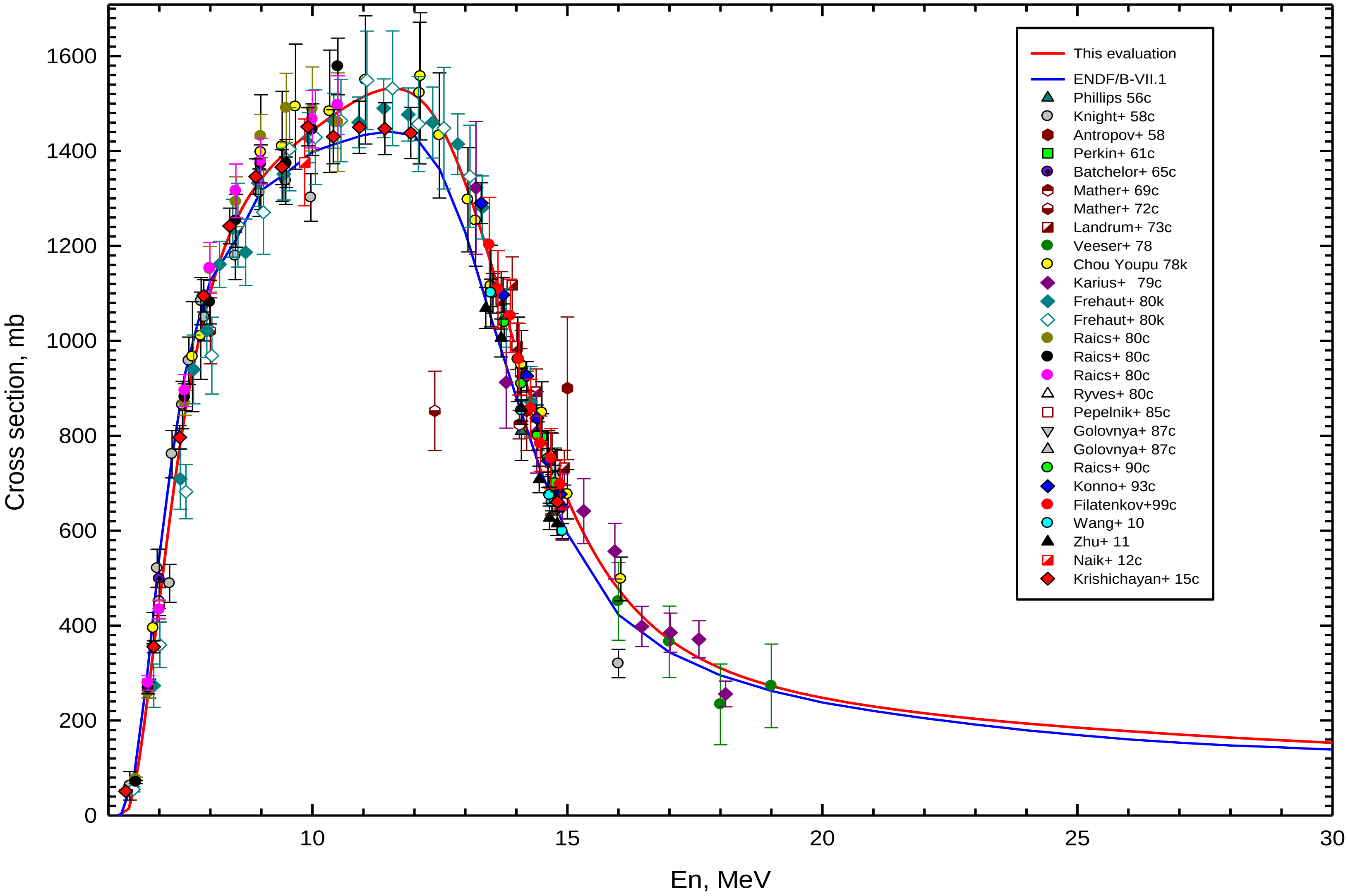

FIG. 1 Evaluated ^{238}U(n,2n)^{237}U reaction excitation function in the energy from threshold to 30 MeV in comparison with equivalent data from ENDF/V-II.1 library and corrected experimental data.

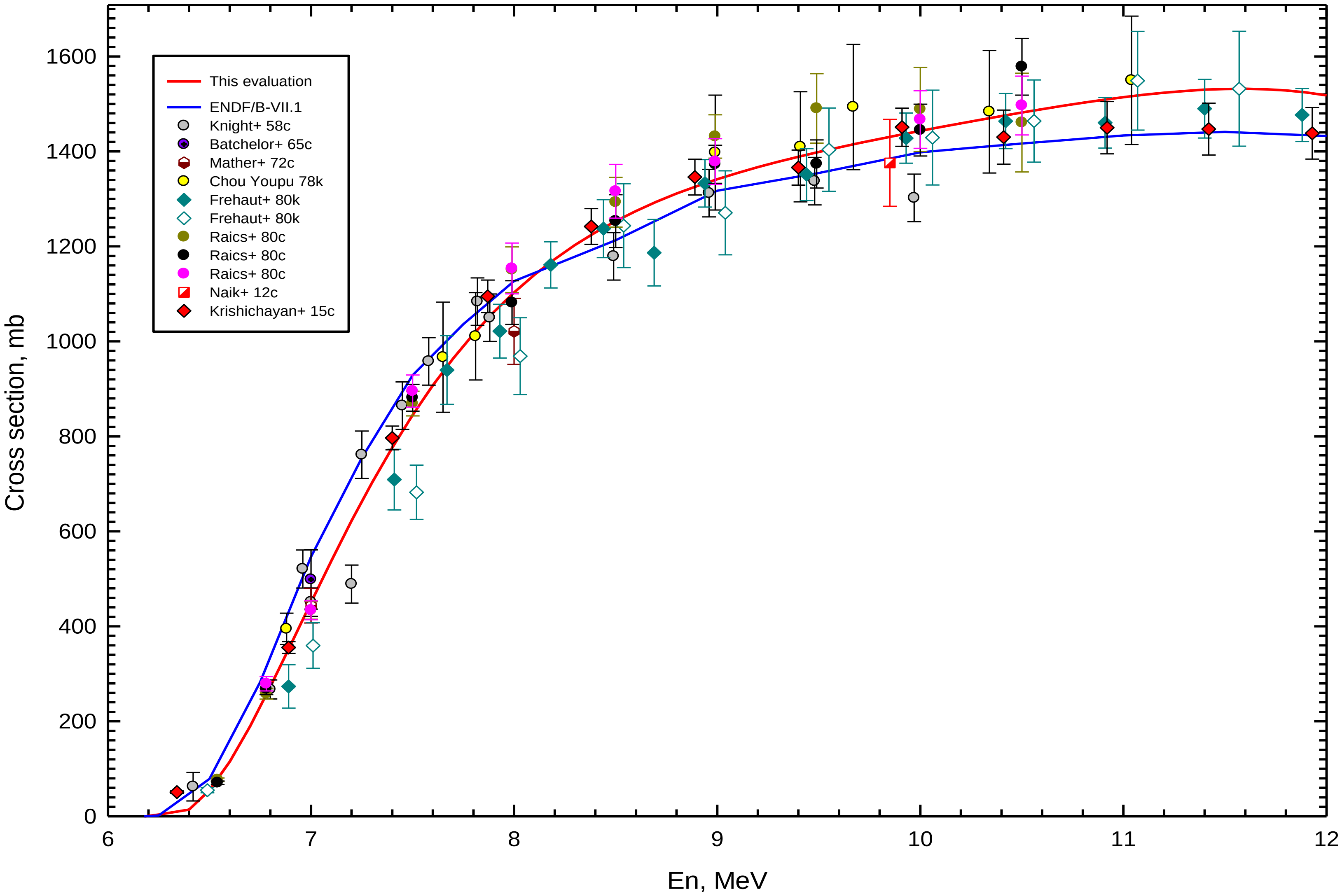

FIG. 2 Evaluated ^{238}U(n,2n)^{237}U reaction excitation function in the energy range from threshold to 12 MeV in comparison with equivalent data from ENDF/V-II.1 library and corrected experimental data.

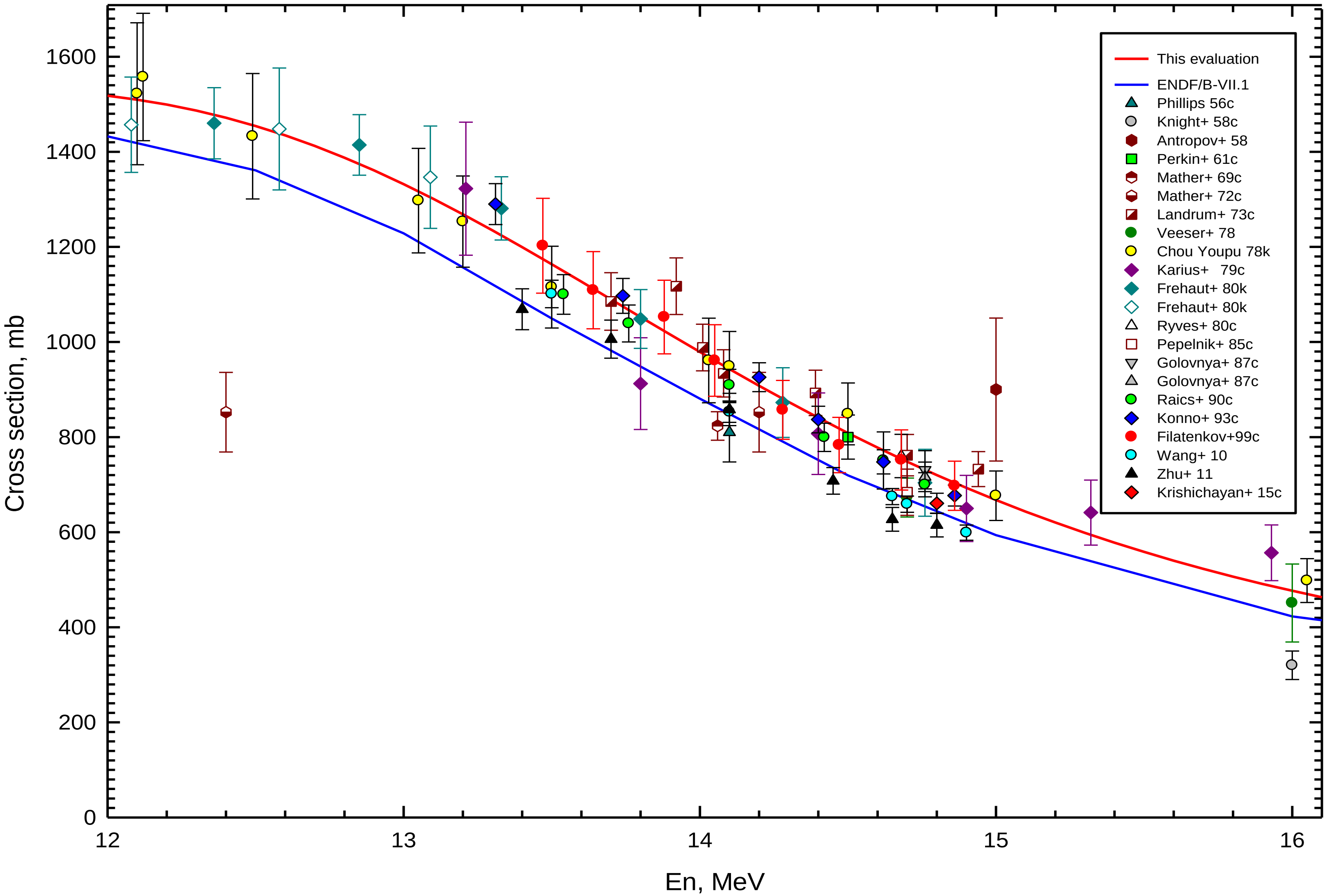

FIG. 3 Evaluated ^{238}U(n,2n)^{237}U reaction excitation function in the energy range 12 - 16 MeV in comparison with equivalent data from ENDF/V-II.1 library and corrected experimental data.

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