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Current status of U^{238} capture cross-section data in the neutron energy region 2 keV - 10 MeV

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A feature of fast reactor development in recent years is the increased importance of large systems with a high degree of fuel dilution and a much smaller neutron spectrum, as a result of which the neutron energy region below 100 keV is becoming particularly important and the need has arisen to know with great accuracy the capture cross-sections of fissionable elements in this energy region. At the same time, known cross-section values for U^{238} are not sufficient for practical needs. The purpose of the present paper is to evaluate the results of recent U^{238} capture cross-section measurements.

The results of U^{238} capture cross-section measurements performed before the Paris conference in 1966 have been collated at the nuclear data centres - Brookhaven, Vienna (IAEA), Saclay and Obninsk - and are reflected in the work of Schmidt [1], in BNL-325 [2] and in newsletters issued by the centres [3, 4]. The results obtained by various authors up to the end of 1966 differ by 15-40% throughout the energy region. Single normalization of the results reduces the discrepancies somewhat, but not substantially, and there is arising an urgent need for new experimental data of sufficient accuracy.

The following criteria were applied in analysing the experimental results: (a) greater weight was attached to new experimental results; (b) the chosen U^{238} capture cross-section value was the one used as reference cross-section for subsequent renormalization; (c) the reference shape of the capture cross-section curve was chosen; (d) data not in agreement with the reference shape of the curve (old data) were rejected.

The weighted mean capture cross-section value for U^{238} at 30 keV was used as reference cross-section value. The experimental data used in obtaining this value are presented in Table 1. An indication is given under "Comments" when renormalization was considered necessary. The weighted mean value was calculated from the formula

$$\bar{\sigma}_{n\gamma} = \frac{\sum_i (\sigma_i/\Delta\sigma_i^2)}{\sum_i \left(\frac{1}{\Delta\sigma_i^2}\right)}; \qquad \Delta\bar{\sigma}_{n\gamma} = \sqrt{\frac{1}{\sum_i \left(\frac{1}{\Delta\sigma_i^2}\right)}}$$

The experimental data obtained by Moxon [17] were used in determining the reference shape of the curve in the region 1-100 keV. These measurements were performed by the time-of-flight method with a flight path of 32.5 m on the Harwell 45-MeV linear electron accelerator, using a Moxon-Rae detector for recording the gamma rays. The cross-section for capture in U^{238} was determined by measuring the efficiency ratio for capture gamma rays in a U^{238} sample and 480-keV gamma rays emitted in the reaction $B^{10}(n,\alpha\gamma)Li^7$ and by subsequently determining the fraction of interactions per incident neutron in the boron sample leading to the emission of 480-keV gamma rays. The efficiency ratio was determined at the 6.7-eV resonance peak, where the fraction of interactions yielding gamma rays per incident neutron approaches unity for the uranium and boron samples and is only very slightly dependent on the resonance parameters.

An undoubted merit of Moxon's work is the fact that the yields of the reactions $U^{238}(n,\gamma)U^{239}$ and $B^{10}(n,\alpha\gamma)Li^7$ are measured simultaneously by the same detector; the measured cross-sections cover the range 0.5-100 keV. The only external source of uncertainties in the experiment is the uncertainty in the cross-section for the reaction $B^{10}(n,\alpha\gamma)Li^7$.

The shape of Moxon's U^{238} capture cross-section curve for the region 1-100 keV is probably the most accurate and the most carefully measured, and it can therefore be recommended for use. The absolute value of the U^{238} capture cross-section at 30 keV is somewhat lower than other known absolute values (Moxon's value is 418 ± 29 mbarn at 30 keV). For normalizing, it is therefore better to take the weighted mean of all existing absolute measurements, including those of Moxon. The original and renormalized mean cross-sections for capture in U^{238} are presented in Table 2.

The data of Menlove and Poenitz [12] for the U^{238} capture cross-section in the energy region 25-500 keV were also renormalized to 466.1 mbarn at 30 keV. Less weight was attached to these data since the U^{235} fission cross-section, measured with the same method of neutron flux determination [23], is 15% lower in the region 300-1500 keV than the values obtained by other authors. Davey [20] points out that a possible reason for the discrepancy between Poenitz's data and those of other authors is incorrect determination of the neutron flux. The original and renormalized data of Menlove and Poenitz are presented in Table 3.

Poenitz [24] has just finished measuring the cross-section ratios $\sigma_{n\gamma}(U^{238})/\sigma_{nf}(U^{235})$ and $\sigma_{n\gamma}(U^{238})/\sigma_{nf}(Pu^{239})$ in the region 130-1400 keV, using neutrons from the reaction $Li^7(p,n)Be^7$. The U^{235} fission cross-section was measured by means of an ionization chamber, while the U^{238} capture cross-section was determined by the activation method. To determine the absolute value, the ratio $\sigma_{n\gamma}(U^{238})/\sigma_{nf}(U^{235})$ was measured at thermal neutron energy. For the ratio $\sigma_{n\gamma}(U^{238})/\sigma_{nf}(Pu^{239})$, the counting efficiencies were determined absolutely to within 2.5%. The experimental cross-section ratio values are presented in Tables 4 and 5.

The cross-sections for U^{235} and Pu^{239} fission were evaluated recently by Davey [25] and Hart [26]. For U^{235} the evaluations did not differ in the energy region 1-700 keV. In the region 0.8-6 MeV, however, Davey's values are 2-5% lower than those of Hart, while in the region 6-10 MeV they are 6-15% lower. In the energy region 130-1400 keV - which is the one of interest to us - the discrepancy is about 2.5% at 900 keV and 1400 keV. These values are presented in Table 4. The cross-section values for Pu^{239} fission are presented in Table 5. Both Hart and Davey have evaluated the ratio $\sigma_{nf}(Pu^{239})/\sigma_{nf}(U^{235})$. There is a discrepancy of the order of 3-9% between their values in the region 1-100 keV and of 3-5% in the region 0.6-5.0 MeV; Davey's data do not include a peak in the region 0.7-2 MeV. In the region 100-600 keV, the two sets of data are in good agreement.

Recent measurements of fission cross-section ratios performed by Pfletschinger [27] and Poenitz [24] confirm Hart's curve in the region 600 keV-2 MeV and are in better agreement with Davey's curve below 100 keV. In calculating the U^{238} capture cross-section we therefore used Hart's values.

The results of the measurements performed by Barry, Bunce and White [19] in the region 127-7600 keV were used as absolute values, without renormalization. This experiment may be considered sufficiently reliable since the method and techniques used for determining the neutron flux (relative to the (n-p) scattering cross-section) were the same as in White's precision measurements of the U^{235} fission cross-section [28]. The original data of Barry, Bunce and White are presented in Table 6.

The U^{238} capture cross-section values evaluated in this way are presented in Table 7. In spite of the difference in the approach to capture cross-section evaluation, including different reference cross-section values, the U^{238} capture cross-section values obtained in the present paper agree to within 2-5% with the results of Davey's recent evaluation [20].

Table 1

Capture cross-sections for U^{238} at 30 keV

Neutron Energy (keV)	$\sigma_{n\gamma}$ (mbarn)	References	Comments
30	370 ± 77	Hanna and Rose [5]	(a)
30,5	461 ± 46	Bilpuch, Weston and Newson [6]	(b)
30 ± 8	470 ± 38	De-Saussure, Weston et al.[4]	(c)
30 ± 7	473 ± 47	Gibbons, Macklin, Miller, and Neiler [8]	(d)
30.1	507 ± 51	Macklin,Gibbons, and Pasma [9]	(e)
30,0	422 ± 34	Belanova et al.[10] with corrections [11, 22]	(f)
30.0±1.5	479 ± 14	Menlove and Poenitz [12]	(g)
30.0	394 ± 85	Bergquist [13]	(h)
30.0±8.0	403 ± 62	Moxon and Chaffey [14]	(i)
30.0±6.0	540 ± 60	Miessner and Arai [15]	(j)
30,0	556 ± 56	Macklin, Lazar and Lyon [16]	(k)
30 ± 8	418 ± 29	Moxon [17]	(l)
30	467 ± 18	Poenitz [18]	(m)
30	466,1 ± 8,6	Present work	(n)

(a) The data are absolute but out of data, and with considerable experimental errors.

(b) Relative measurements at 3 - 217 keV. The shape of the curve does not agree with Moxon's measurements [17], so that the data were normalized to Barry's data [19] at 130 - 217 keV by multiplying them by the coefficient 0,96.

(c) Absolute measurements at 30 keV and 64 keV. The ratio $\sigma_{n\gamma}(U^{238})/(\sigma_{nf}+\sigma_{n\gamma})(U^{235})$ was measured (= 0,150 ± 0,012 at 30 keV.) Davey [20] renormalized the results using $\sigma_{nf}(U^{235}) = 2,28$ barn and $\alpha(U^{235}) = 0,373$, which is the weighted mean of the values of De Saussure et al.

(0,372 ± 0,026 [7] and Hopkins and Diven (0,376 ± 0,036 [21]). Then $(\sigma_{nf} + \sigma_{n\gamma})$ (U^{235}) = 3,13.

(d) Absolute measurements at 30 keV and 65 keV. The absorption cross-section for indium at 30 keV (763 mbarn) was used as a standard.

(e) Absolute measurements in the range 8 - 58 keV. The data for $\sigma_{n\gamma}(U^{238})$ were obtained by assuming that the tantalum capture cross-section was σ(Ta) = 8600 exp (- 0,697 ln E) mbarn. Davey [20] renormalized the original U^{238} capture cross-section data using the following formula for the tantalum capture cross-section: σ(Ta) = 8600 exp (-0,720 ln E) mbarn. The original value was $\sigma_{n\gamma}$ (U^{238}) = 549 ± 55 mbarn; the renormalized value is $\sigma_{n\gamma}$ (U^{238}) = 507 ± 51 mbarn at 30,1 keV.

(f) Absolute measurements at 23 meV. Poenitz and Miller [11, 22] reviewed the results of Belanova et al. [10]. Using resonance parameters taken from various publications, they employed the Monte Carlo method to calculate corrections for resonance self-shielding in the uranium sample. Introducing these corrections, they obtained a value of 495 ± 40 mbarn for the U^{238} capture cross-section at 23 keV instead of the value of 412 ± 18 mbarn reported by Belanova. The shape of Moxon's curve [17] was used in extrapolating from 22,8 keV to 30 keV.

(g) Absolute measurements at 30 keV.

(h) Relative measurements in the range 18 - 300 keV. The U^{238} capture cross-section data were normalized to $\sigma_{n\gamma}$(Ag) = 1,185 barn at 24 keV. The original value of the U^{238} capture cross-section at 33 keV was 325 ± 50 mbarn. The results were inaccurate and were therefore renormalized to the data of Barry et al. [19] at 125 - 300 keV. The mean value of the renormalization coefficient was 1,28 ± 0,2.

(i) Normalization as in [17]

(j) The measurements were made by bombarding a block of uranium with 30-keV neutrons. The resulting U^{238} capture cross-section value was not measured directly; moreover, a more carefully calculated correction for resonance self-shielding is necessary.

(k) Absolute measurements at 25 keV gave the result $\sigma_{n\gamma}(U^{238})$ = 610 ± 61 mbarn, as compared with a measured cross-section for iodine of 820 ± 60 mbarn at 25 keV. The shape of Moxon's curve [17] was used in extrapolating to 30 keV.

(l) Absolute measurements with normalization to the resonance peak at 6,7 eV.

(m) The ratio $\sigma_{n\gamma}(U^{238})/\sigma_{nf}(U^{235})$ was measured at 30, 130, 500 and 900 keV. At 30 keV the value of the ratio was 0,205 ± 0,008, while $\sigma_{nf}(U^{235})$ = 2,28 barn.

(n) Weighted mean capture cross-section value obtained from all the above data.

Table 2

Original and renormalized data for $\sigma_{n\gamma}$ (U^{238})
(Moxon [17])

Average neutron energy (keV)	Measured $\sigma_{n\gamma} U^{238}$ (mbarn)	Renormalized data, $\sigma_{n\gamma}$ (U^{238}) x 1.11 (mbarn)
0.55	3845 ± 126	4290 (a)
0.65	2995 ± 105	3340
0.75	1712 ± 78	1910
0.85	2782 ± 102	3104
0.95	3121 ± 109	3480
1.5	1772 ± 75	1980
2.5	1372 ± 64	1530
3.5	1156 ± 59	1290
4.5	921 ± 52	1028
5.5	856 ± 54	955
6.5	825 ± 49	920
7.5	760 ± 46	848
8.5	701 ± 45	782
9.5	701 ± 43	782
15	594 ± 35	662
25	460 ± 28	513
30	418 ± 29	466
35	380 ± 33	424
45	351 ± 22	392
55	305 ± 18	340
65	253 ± 15	282
75	208 ± 13	232
85	192 ± 16	214
95	183 ± 15	204

(a) The data were renormalized to the weighted mean value of 466,1 mbarn at 30 keV.

Table 3

Original and renormalized data for $\sigma_{n\gamma}(U^{238})$
(Menlove and Poenitz [12])

Average neutron energy (keV)	Measured $\sigma_{n\gamma}(U^{238})$ (mbarn)	Renormalized data $\sigma_{n\gamma}(U^{238})$ (mbarn)	
24.4 ± 0.9	516 ± 27	503	(a)
30.0 ± 1.5	479 ± 14	466	
43.8 ± 1.3	404 ± 19	394	
63.3 ± 0.7	302 ± 14	294	
97.3 ± 2.8	201 ± 11	196	
157.0 ± 3.0	153 ± 9	149	
264.0 ± 3.0	126 ± 9	123	
373.0 ±12.0	123 ± 11	120	
503.0 ±12.0	110 ± 10	107	

(a) The data were renormalized to the weighted mean value of 466,1 mbarn (at 30 keV).

Table 4

Data for the ratio $\dfrac{\sigma_{n\gamma}(U^{238})}{\sigma_{nf}(U^{235})}$

(Poenitz [24])

Neutron energy (keV)	Measured $\dfrac{\sigma_{n\gamma}(U^{238})}{\sigma_{nf}(U^{235})}$	σ_{nf} (U^{235}) Hart [26] (barn)	σ_{nf} (U^{235}) Davey [25] (barn)	$\sigma_{n\gamma}$ (U^{238}) values derived using Hart values (mbarn)
30(a)	0.205 ± 0.008	2.28(b)	2.28	467 ± 27
130	0.126 ± 0.006	1.55	1.54	195 ± 13
150	0.126 ± 0.006	1.50	1.50	189 ± 13
250	0.114 ± 0.004	1.35	1.35	154 ± 8
300	0.103 ± 0.003	1.30	1.30	134 ± 7
400	0.104 ± 0.003	1.25	1.25	130 ± 6
500	0.111 ± 0.003	1.17	1.17	130 ± 6
600	0.122 ± 0.004	1.135	1.135	138 ± 7
700	0.133 ± 0.004	1.135	1.14	151 ± 7
900	0.124 ± 0.004	1.18	1.21	141 ± 8
1200	0.097 ± 0.004	1.23	1.22	119 ± 7
1250	0.092 ± 0.004	1.239	1.22	114 ± 6
1400	0.074 ± 0.003	1.25	1.22	92.5 ±6

(a) Value taken from [18]

(b) The error in the evaluated curve is thought to be 2 %.

Table 5

Data for the ratio $\frac{\sigma_{n\gamma}(U^{238})}{\sigma_{nf}(Pu^{239})}$

(Poenitz [24])

Neutron energy, (keV)	Measured $\frac{\sigma_{n\gamma}(U^{238})}{\sigma_{nf}(Pu^{239})}$	$\sigma_{nf}(P^{239})$ Hart [26] (barn)	σ_{nf} (P^{239}) Davey [25] (barn)	$\sigma_{n\gamma}(U^{238})$ values derived using Hart values (mbarn)
250	0.095±0.004	1.53(a)	1.53	145 ± 9
400	0.082±0.003	1.54	1.55	126 ± 7
500	0.082±0.002	1.56	1.57	128 ± 6
700	0.087±0.003	1.66	1.61	145 ± 8
900	0.086±0.003	1.71	1.74	147 ± 8
1400	0.048±0.002	1.93	1.80	92.5 ± 6

(a) The error in the evaluated curve is thought to be 2 %.

Table 6

Original and renormalized data of Barry, Bunce and White [19]

Neutron energy (keV)	measured $\sigma_{nf}(U^{238})$ (mbarn)	$\sigma_{nf}(U^{235})$ Huges and Schwarz (barn)	$\sigma_{nf}(U^{235})$ Davey [4] (barn)	$\sigma_{nf}(U^{235})$ Hart [3] (barn)	$\sigma_{nf}(U^{238})$ values renormalized using Hart values (mbarn)
127 ± 20	218±11				
160 ± 24	200± 8				
207 ± 23	158± 6				
312 ± 22	141± 6				
404 ± 22	126± 5				
505 ± 22	128± 5				
810 ±110	147± 8				
1060 ±105	149± 7				
1300 ±105	130± 7	1.28 (a)	1.22	1.24	126
1750 ±105	69± 4	1.30	1.27	1.283	68
3000 ±115	27± 2	1.30	1.18	1.23	26
5000 ±200	11.0±1.5	1.15	1.04	1.09	10.4
7600 ±200	6.4±2.0	1.70	1.61	1.73	6.5

(a) $\sigma_{nf}(U^{235})$ was used by Barry et al. to derive $\sigma_{nf}(U^{238})$ at energies higher than 1 MeV.

Table 7

Evaluated capture cross-sections for U^{238}

Lethargy	Neutron energy (keV)	$\sigma_{n\gamma}(U^{238})$ present work (mbarn)
8.5	2.03	1650
8.4	2.25	1570
8.3	2.49	1525
8.2	2.75	1455
8.1	3.04	1390
8.0	3.35	1310
7.9	3.71	1240
7.8	4.10	1165
7.7	4.53	1075
7.6	5.00	1000
7.5	5.53	955
7.4	6.11	925
7.3	6.76	890
7.2	7.47	845
7.1	8.25	805
7.0	9.12	780
6.9	10.1	755
6.8	11.1	730
6.7	12.3	705
6.6	13.6	676
6.5	15.0	650
6.4	16.6	620
6.3	18.4	595
6.2	20.3	567
6.1	22.4	535
6.0	24.8	515
5.9	27.4	486
5.8	30.3	465
5.7	33.5	438
5.6	37.0	416

Table 7 (continued)

Lethargy	Neutron energy (keV)	$\sigma_{n\gamma}$ (U238) present work (mbarn)
5.5	40.9	403
5.4	45.2	390
5.3	49.9	370
5.2	55.2	343
5.1	61.0	305
5.0	67.4	270
4.9	74.5	240
4.8	82.3	218
4.7	91.0	210
4.6	101.0	205
4.5	111.0	202
4.4	123.0	197
4.3	136.0	190
4.2	150.0	184
4.1	166.0	175
4.0	183.0	165
3.9	202.0	157
3.8	224.0	150
3.7	247.0	145
3.6	273.0	139
3.5	302	133
3.4	334	131
3.3	369	128
3.2	408	127
3.1	450	128
3.0	0.497	129
2.9	0.550	133
2.8	0.608	141
2.7	0.672	146
2.6	0.742	148
2.5	0.821	148
2.4	0.907	147
2.3	1.000	145
2.2	1.11	135

Table 7 (continued)

Lethargy	Neutron energy (keV)	$\sigma_{n\gamma}(U^{238})$ present work (mbarn)
2.1	1.22	117
2.0	1.35	98
1.9	1.50	83
1.8	1.65	73
1.7	1.83	65
1.6	2.02	58
1.5	2.23	50
1.4	2.47	43
1.3	2.73	32
1.2	3.01	26
1.1	3.33	22
1.0	3.68	18
0.9	4.07	16
0.8	4.49	13
0.7	4.97	11
0.6	5.49	9
0.5	6.07	7.8
0.4	6.70	7.0
0.3	7.41	6.2
0.2	8.19	5.6
0.1	9.05	5.0
0.0	10.00	4.5

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