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FISSION CROSS-SECTION NORMALIZATION PROBLEMS

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INTRODUCTION

Before discussing the fission cross-section normalization problems it is useful to first recall their origin(s). What the measuring society wants to achieve is to determine the ^{235}U(n,f) cross-section with 1% accuracy from almost 0 to 20 MeV neutron energy. Two major problems make this hard to realise: First no single neutron source covers the whole energy region of interest, and secondly no single neutron flux detector does so. Hence one has to work with partial measurements which have to be joined together, causing normalization and cross-normalization problems. This is especially true for the shape measurements, in which the shape of the ^{235}U(n,f) cross-section is measured relative to that of a reference cross-section (^{10}B(n,α), ^{6}Li(n,α), H(n,n'), ...). Such measurements have to be normalized to absolute fission cross-section values, which, however, are only available for a limited number of neutron energies (mainly in the thermal region and between 0.1 and 14 MeV). Ideally all fission cross-section measurements should be normalized in the thermal region, since here one combines at the same time large cross-section values and an isotropic emission of the fission fragments with the availability of very high neutron fluxes. So thin samples can be used, which reduces several experimental uncertainties such as self-absorption, scattering, energy loss etc. As a consequence of these excellent experimental conditions, the (absolute) thermal fission cross-section of ^{235}U can be determined with an accuracy of $\leq$ 1% (e.g. ref. 1). At higher neutron energies ($\geq$100 keV), the accuracy of the absolute fission cross-section measurements is typically 2-3%, with the exception of the 14 MeV region where the favourable kinematics of the ^{3}H(d,α)n neutron source enables a 1.5% accuracy (ref. 2). Another reason for the high accuracy achieved in (ref. 2) was the thorough check of samples and detectors with a thermal neutron beam. We strongly recommend such a procedure for all absolute measurements in the high energy region. A thermal neutron beam is indeed a powerful tool for checking targets and detectors and for resolving discrepancies between different targets (detectors). A typical example for the latter application can be found in (ref. 3).

THE USE OF NORMALIZATION INTEGRALS

For various reasons it is often not possible to normalize a fission cross-section shape measurement in the thermal region. This is illustrated in Table 1. Out of 11 measurements performed before 1970, only one (ref. 2) is directly, and three others (refs. 4, 7, 8) are indirectly normalized in the thermal region. Moreover, some of the normalization methods reported

Table 1 COMPARISON OF THE NORMALIZATION METHODS APPLIED IN THE OLDER σ_f MEASUREMENTS OF ^{235}U (before 1970).

SHORE and SAILOR (4)	σ_f normalized to the data of LEONARD (unpublished) in the region 0.1-0.4 eV which are normalized at 0.0253 eV to (582±10) barn (two-step normalization).
MICHAUDON (5)	σ_f normalized to $\int_{8\ \text{eV}}^{10\ \text{eV}} \sigma_f(E)dE = 208.47$ barn.eV obtained by SHORE and SAILOR.
MICHAUDON et al. (6)	σ_f normalized to $\int_{0.4\ \text{eV}}^{1.3\ \text{eV}} \sigma_f(E)dE$ obtained by SHORE and SAILOR.
IGNATIEV et al. (7)	σ_f deduced from measurements of σ_t and η, assuming $\sigma_s = \frac{22}{(E+2.3)^2} + 11$ (σ_s in barn, E in eV) and normalized to η=2.07 at 0.0253 eV. Indirect method.
RYABOV et al. (8)	σ_f normalized to $\sigma_f{}^{\circ}$= (582±6 barn). The lowest data point given by the author is 0.15 eV. The error introduced by the normalization is 5 per cent.
BOWMAN et al. (9)	σ_f normalized at 0.0253 eV to the least squares value of (577.1±0.9)barn reported in BNL 325, suppl.2.
BROOKS et al. (10)	σ_f calculated via $\eta = \frac{\bar{\nu}\sigma_f}{\sigma_t - \sigma_s}$ and normalized at 0.06 eV by putting η= 2.084 assuming $\bar{\nu}$= 2.42. Indirect method
MOSTOVAYA et al. (11)	σ_f normalized to the averaged fission cross-section curve given in BNL 325 in the region 0.8-0.9 eV.
DE SAUSSURE et al. (12)	σ_f normalized by making the resonance integral $\int_{0.45\ \text{eV}}^{10\ \text{eV}} \sigma_f(E)\frac{dE}{E}$ equal to 127.9 barn (average of the results of SHORE and SAILOR and BOWMAN).
CAO et al. (13)	σ_f normalized to $\int_{8\ \text{eV}}^{10\ \text{eV}} \sigma_f(E)dE = 208.47$ barn eV obtained by SHORE and SAILOR.
BLONS et al. (14)	σ_f normalized to the integrated fission cross-section from 60-200 eV from MICHAUDON. Indirect normalization.

Table 2 COMPARISON OF FLUX DETERMINATION AND NORMALIZATION METHODS APPLIED IN THE QUOTED σ_f-MEASUREMENTS OF ^{235}U (AFTER 1970)

Authors	Neutron Flux Determination	Normalization	Corresponding $\sigma_f{}^*$ (barn)
De Saussure (15)	^{10}B(n,α)	$\int_{7.8\ \text{eV}}^{11\ \text{eV}} \sigma_f(E)dE = 240$ barn.eV (Deruytter and Wagemans, 16)	587.6
Deruytter and Wagemans (16)	^{10}B(n,α)	$\int_{0.02060\ \text{eV}}^{0.06239\ \text{eV}} \sigma_f(E)dE = 19.27 \pm 0.08$ barn.eV (Deruytter et al., 1)	587.9
Silver et al. (17)	^{10}B(n,α)	σ_f of De Saussure et al. (12) between 100 and 200 eV	577.1
Lemley et al. (18)	^{6}Li(n,α)	$\sigma\ [^{6}\text{Li}(n,\alpha)]$	
Blons (19)	^{10}B(n,α)	$\int_{100\ \text{eV}}^{200\ \text{eV}} \sigma_f(E)dE = 2103$ barn.eV (De Saussure, 15)	587.6
Gayther et al. (20)	Calibrated boron-vaseline plug	$\bar{\sigma}_f = 2.349$ barn in the interval 10-30 keV (Soverby, 32)	580.2
Perez et al. (21)	^{10}B(n,α)	$\int_{100\ \text{eV}}^{200\ \text{eV}} \sigma_f(E)dE = 2103$ barn.eV (De Saussure, 15)	587.6
Perez et al. (22)	^{10}B(n,α)	$\int_{2\ \text{keV}}^{10\ \text{keV}} \sigma_f(E)dE = 31.643$ barn keV (Perez et al., 21)	
Czirr et al. (23)	^{6}Li(n,α)	$\sigma_f = 585.4$ barn	585.4
Wasson (24)	^{6}Li(n,α)	$\int_{7.8\ \text{eV}}^{11\ \text{eV}} \sigma_f(E)dE = 238.4$ barn eV	(583.5)
Gwin et al. (25)	^{10}B(n,α)	σ_f of ENDF-B III between 0.02-0.4 eV	580.2
Wagemans and Deruytter (26)	^{10}B(n,α) $[^{6}$Li(n,α)]	$\int_{7.8\ \text{eV}}^{11\ \text{eV}} \sigma_f(E)dE = 240$ barn.eV	587.6
Czirr et al. (27)	^{6}Li(n,α)	$\int_{0.02\ \text{eV}}^{0.1\ \text{eV}} \sigma_f(E)dE$ of Leonard (33)	585.4
Muradjan et al. (28)	^{10}B(n,α)	$\int_{100\ \text{eV}}^{1000\ \text{eV}} \sigma_f(E)dE = 12\,209$ barn.eV	
Moore et al. (29)	^{6}Li(n,α)	$\int_{7.8\ \text{eV}}^{11\ \text{eV}} \sigma_f(E)dE = 241.2$ barn.eV	583.5
Wagemans et al. (30)	^{10}B(n,α) ^{6}Li(n,α) *	$\int_{0.0206\ \text{eV}}^{0.06239\ \text{eV}} \sigma_f(E)dE = 19.26 \pm 0.08$ barn.eV (Deruytter et al., 1)	587.6
Corvi et al. (31)	^{6}Li(n,α)	$\int_{7.8\ \text{eV}}^{11\ \text{eV}} \sigma_f(E)dE = 241.2$ barn eV	583.5
This work	^{6}Li(n,α)	$\int_{0.0206\ \text{eV}}^{0.06239\ \text{eV}} \sigma_f(E)dE = 19.26$ barn eV	587.6

* up to 200 eV

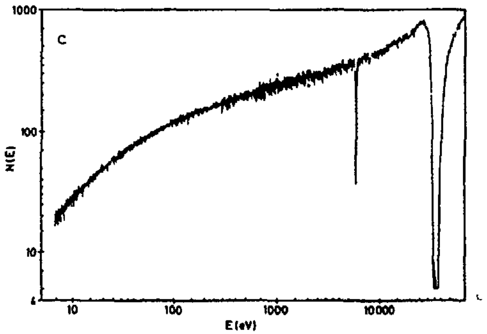
in this table are peculiar and/or unreliable. Also for the more recent measurements reported in Table 2, only 6 out of 18 measurements are directly normalized in the thermal region.

For measurements not reaching the thermal region, the introduction of accurate secondary normalization integrals is very useful. Such integrals may also provide a link between absolute cross-section measurements at thermal and at higher neutron energies. In this context we proposed some years ago the fission integral $I_1 = \int_{7.8\,\mathrm{eV}}^{11\,\mathrm{eV}} \sigma_f(E)dE$, which is a rather favourable choice since it contains a large resonance, yielding a high counting rate and the signal to background ratio is good. Furthermore, since the cross-section values at its limits are small, this integral is not sensitive to bad resolution and small timing errors.

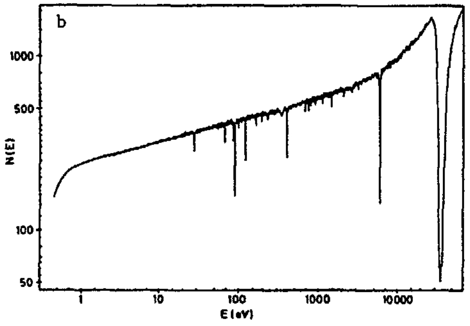
Another integral which may play a role as a secondary normalization integral is $I_2 = \int_{0.1\,\mathrm{keV}}^{1.0\,\mathrm{keV}} \sigma_f(E)dE$, which should be especially useful for measurements not coming down to the eV-region.

Anyhow, these integrals should not be used blindly as is illustrated in Fig. 1. This figure shows the $^{10}B(n,\alpha)$ counting rate (for a constant time-of-flight channel width) in arbitrary units, as a function of the neutron energy (a) without an overlap filter, (b) with a cadmium overlap filter, (c) with a boron overlap filter. These data were obtained at the Geel linear accelerator GELINA. From this figure it is obvious that the neutron energy region from 100 to 1000 eV (I_2) is not suitable for normalization purposes for measurements using a Cd overlap filter, because of the presence of Cd transmission dips in this region. For measurements using a ^{10}B-overlap filter, the neutron flux in the eV-region is strongly reduced. In the example shown in Fig. 1c, about 85% of the 10eV-neutrons are absorbed in the overlap filter. So before normalizing such a measurement via I_1, one has to check the signal to background ratio in the 10eV energy region.

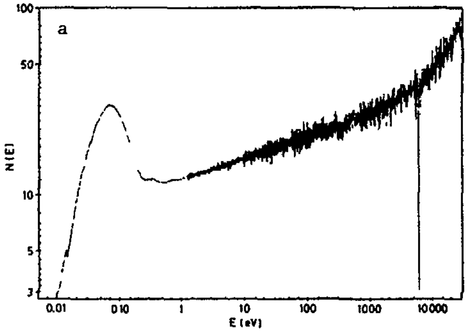
It is inherent to a good normalization that the final result does not depend on the normalization procedure used. E.g. normalizations via the integral I_1 or I_2 should be consistent. The large spread on the I_2-values reported in the literature (~7%) is problematic in this respect. One of the origins of these discrepancies is certainly the fact that most of the I_2-values have been obtained via an indirect normalization procedure. Hence the first aim of a recently started series of experiments at the Geel linear accelerator was to determine I_1 and I_2 in one single experiment, directly normalized



^{10}B overlap filter



Cd overlap filter



no overlap filter

Fig. 1 : $^{10}B(n,\alpha)$ counting-rates as a function of the neutron energy (a) without overlap filter (b) with a cadmium overlap filter (c) with a boron overlap filter.

in the thermal region. The second goal was to cover the neutron energy region from 0.02 eV up to 30 keV in that same experiment, thus realizing a link between the thermal region and absolute measurements in the lowest part of the higher energy region (e.g. Szabo and Marquette, ref. 34).

EXPERIMENTAL CONDITIONS

No such measurement is available until now. The reason could be the rather exotic experimental conditions needed. One has indeed to cover a very large dynamic range with strongly different time of flight resolution requirements. At the Geel linear accelerator these requirements could be fulfilled by using a 4 ns time-coder with two million channels and an "accordeon" system (i.e. a variable t.o.f. channel width). The linac was operated at a repetition frequency of 100 Hz and a 4 ns pulse width. The $^{235}U(n,f)$-fragments and the $^{6}Li(n,\alpha)t$ reaction products were simultaneously detected with surface barrier detectors from the same position in the neutron beam. In all our previous experiments a low detection geometry with the surface barrier detectors outside the neutron beam was used. In these experiments an almost 2π-geometry was realized by sandwiching the back-to-back ^{235}U and ^{6}Li foils between both surface barrier detectors; the whole system was placed into the collimated neutron beam. The thickness of the ^{6}LiF-target was 88 $\mu g/cm^2$. For the fission reaction, two independent measurements were performed respectively with a 100$\mu g/cm^2$ and a 500$\mu g/cm^2$ evaporated $^{235}UF_4$-layer.

The raw data are shown in Fig. 2. Typical $^{235}U(n,f)$ and $^{6}Li(n,\alpha)t$ counting-rate spectra (per 8 ns t.o.f. channel) are shown in the neutron energy region from 0.01eV up to 50 keV. The background has been determined with the black resonance technique and the data reduction was done as explained previously (e.g. ref. 30). The background was very low. A few typical background values for the $^{235}U(n,f)$ measurement with the 100$\mu g/cm^2$ sample are given within brackets: 0.0253eV (0.2%); 8.78eV resonance (0.15%); 11eV valley (5%); 30 keV (2%); 0.1 - 1 keV (between 1 and 5%). The fission cross-section was normalized to the fission integral $\int_{0.0206\,eV}^{0.06239\,eV} \sigma_f(E)dE = 19.26 \pm 0.08$ barn.eV as determined by Deruytter et al. (1). A check via a normalization procedure using a least square fit in the thermal region yielded a consistent normalization factor. Preliminary numerical values are given in Tables 3 - 6. These results will be discussed in the following chapter.

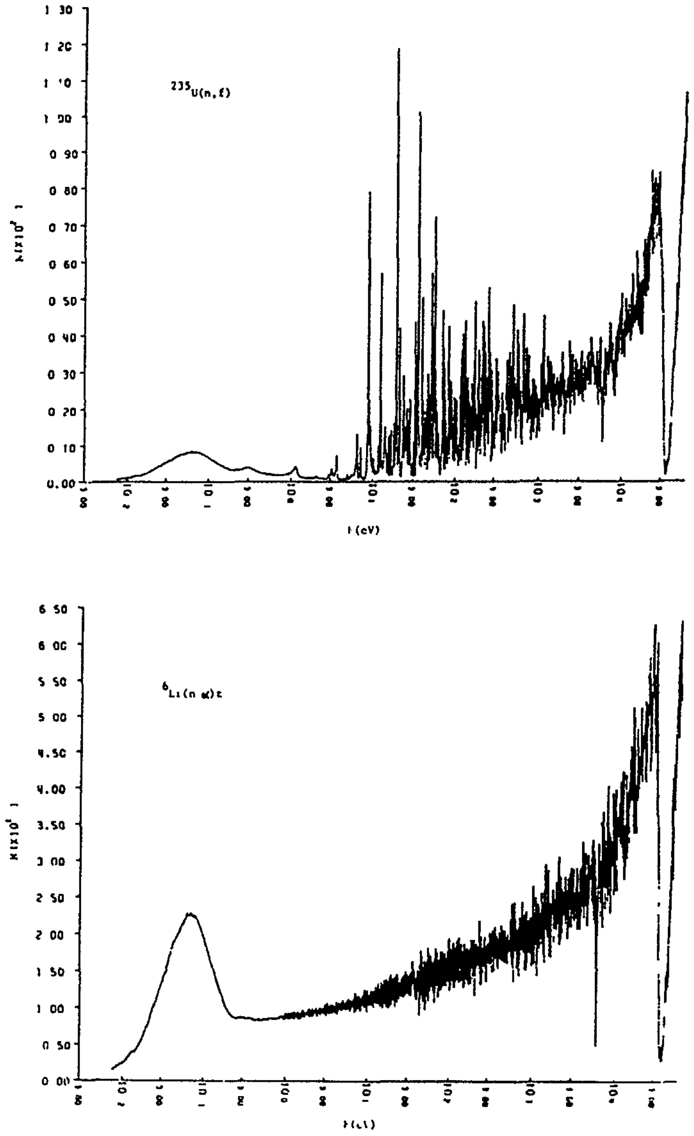


Fig. 2 $^{235}U(n,f)$ and $^{6}Li(n,\alpha)t$ counting-rate spectra reduced to a constant 8 ns t.o.f. channel width

83 RESULTS AND DISCUSSION

A first group of results is given in Table 3, where the absolute fission cross-section data of Szabo and Marquette (34) are compared with the corresponding average fission cross-section values obtained from the present experiments. Obviously, both measurements agree within the experimental errors.

In Table 4 the average fission cross-sections obtained in the present measurements for a series of energy intervals are compared with other results. A first observation is that the results of the measurements with a 100 and with a 500 $\mu g/cm^2$ $^{235}UF_4$ sample are compatible. A second and more puzzling observation is that for intervals containing strong resonances (= high $\bar{\sigma}_f$-values) the present results are systematically lower than in most of the other data sets. This is especially true for the 10 - 20; 30 - 40 and 50 - 60 eV intervals. However, for intervals with $\bar{\sigma}_f \leqslant 30$ barn, the present results are quite plausible... A last observation is that the measurements relative to a $^6Li(n,\alpha)t$ flux monitor generally yield lower $\bar{\sigma}_f$-values than those made relative to a $^{10}B(n,\alpha)$-flux monitor.

In Table 5 a comparison is made of the fission integrals I_1 and I_2 relative to their thermal normalization values. For measurements with an indirect thermal calibration the σ_f°-value has been put within brackets. Also the I_1 and I_2-values obtained in the present experiments are somewhat surprizing: whilst the I_1/σ_f°-values are rather low, the I_2/σ_f°-values are in perfect agreement with the values of Gwin et al. (25) and Gzirr et al. (27), which are the only other measurements in which the I_2-integral was directly normalized in the thermal region. This confirms our previous observation, since the integral I_1 corresponds to a $\bar{\sigma}_f$-value of more than 70, whilst for I_2 $\bar{\sigma}_f \approx 13$ barn.

In Table 6 finally we compare the original values of the integrals I_1 and I_2 together with the neutron flux monitor used. By calculating the ratio of both integrals, the normalization constant is removed. Hence with consistent data sets only small fluctuations should occur. However, Table 6 reveals differences up to 10% between I_2/I_1-values. Another important observation is the apparent correlation between I_2/I_1 and the neutron flux monitor used. Measurements using a ^{10}B flux monitor systematically yield higher I_2/I_1-values than measurements relative to a ^6Li flux monitor, the exception being the present results which should, however, be considered cautiously in view of the surprisingly low I_1-values. Moreover, these low I_2/I_1-values in the Li-case are mainly a consequence of low I_2-values.

Table 3 Comparison of the absolute σ_f-data of Szabo and Marquette (ref.34) with the corresponding (preliminary) $\bar{\sigma}_f$-values of the present work.

E_n (keV)	σ_f (barn) (ref.34)	$\bar{\sigma}_f$ (barn)*
11.5 ± 3	2.76 ± 0.09	2.85
15.0 ± 3	2.50 ± 0.07	2.48
17.5 ± 3.5	2.15 ± 0.09	2.25
22.5 ± 2.5	2.20 ± 0.06	2.17
27 ± 3.5	2.10 ± 0.08	2.03

* average of the measurements with 100 and with 500 $\mu g/cm^2$

TABLE 4 THE (PRELIMINARY) AVERAGE CROSS SECTIONS (BARN) OBTAINED IN OUR MEASUREMENTS COMPARED WITH OTHER RECENT MEASUREMENTS, MAINTAINING THEIR ORIGINAL NORMALIZATION

INTERVAL REF.	.02-.1 eV	.1-.5 eV	5-1. eV	1-10 eV	10-20 eV	20-30 eV	30-40 eV	40-50 eV	50-60 eV	60-100 eV	.1-.2 keV	.2-.3 keV	.3-1. keV	1-10 keV	10-20 keV	20-30 keV
De Saussure (15)				40.14	52.95	36.27	57.06	33.33	61.99	24.34	21.03	20.86	11.58			
Ryabov (8)					46.09	35 05	52 12	32 21	51.10	24.23	21.39	20.83	11.69	4.41	2.98	2.51
Silver (17)										25.75	21.03	20.61	11.59	3.99	2.77	2.37
Lemley (18)										24.05	20.90	20.15	11.09	3.99	2.34	2.10
Blons (19)						34.28	57.31	34.00	64.47	25.15	21.03	20.77	11.71	4.38	2.54	2.20
Gayther (20)															2.53	2.17
Perez (21)					50.57	36 14	55 65	33 38	61.81	24.55	21.03	20.92	11.69	4.35		
Perez (22)															2.53	2.18
Czirr (23)											19.9	19.8	10.71		2.35	2.17
Wasson (24)											20.3	19.9			2.48	2.10
Gwin (25)	379.1	157 2	61.54							23.58	20.47	19.74	11.08	4.08	2.46	2.11
Wagemans (A)	384.1	159.1	60.64	40.64	52 62	38.08	59.02	34 57	64.59	25.40	21.25	20.91	11.53	4.41	2.64	2.22
(26) (B)											21.11	20.71	11.37	4.29	2.54	2.14
Czirr (27)	381.7	159	61.03							24.02	20.23	19.93	10.76			
Muradjan (28)											20.79	19.94	11.64	4.24	2.51	2.17
Moore (29)					52.35	37 36	55 19	33 11	60 14	23.85	20.73	19.53	10.80	3.87	2.52	
Wagemans (C)	384 7	162.9	63.44	41.58	53.64	37 76	58.19	33.66	63.73	25.24	21.43	21.29	11.81	4.31	2.59	2.22
(30) (B)											21.29	21.09	11.65	4.19	2.49	2.14
Biriukov (35)											21.88	20.87	11.62	4.29	2.49	2.09
Corvi (31)											20.37	20.16	11.19	4.16	2.46	2.10
This work (D)	384.0	162.7	66.3	41.1	50.7	37.2	52 8	33.3	59.0	23.9	20.4	20.0	11.3	4.27	2 56	2.10
(prelim.) (E)	384.7	161 4	65 4	40.5	49 8	36.9	52.5	32.9	58.4	24.2	19.8	19 6	11.2	4.24	2.47	2.10
ENDF-BV	382.4	159.7	62 4	41 93	54 54	38.07	56.73	34 35	63.01	23.95	20 54	20 15	11.22	4.20	2.48	2.12

(A) ORIGINAL DATA

(B) CORRECTED FOR NON 1/V 10B(n,α)-SHAPE

(C) REVISED DATA (SEE LIST OF REFERENCES)

(D) 500 $\mu g/cm^2$ ^{235}U target

(E) 100 $\mu g/cm^2$ ^{235}U target

Table 5 Comparison of the integrals I_1(7.8 eV, 11 eV) and I_2 (0.1 keV, 1keV) relative to their thermal normalization values.

Reference	σ_f°(barn)	I_1(barn . eV)	I_1/σ_f° (eV)	I_2(barn.keV)	I_2/σ_f°(eV)
Shore & Sailor (4)	582	229.4	0.394		
Ryabov et al.(8)	582	217.8	0.374		
Michaudon et al. (6)	(582)	232.6 238.1	0.399 0.409		
Brooks et al. (10)	-	215.1	-		
Bowman et al. (9)	577.1	246.7	0.427		
De Saussure et al. (12)	(577.1)	236.7	0.410		
Mostovaya et al. (11)	-	255.6	-		
Cao et al. (13)	(582)	226.6	0.389		
Deruytter & Wagemans (16)	587.9	240.2	0.409		
Gwin et al. (25)	580.2	234.6	0.404	11.79	20.32
Czirr et al. (27)	585.4	244.7	0.418	11.54	19.71
Wagemans & Deruytter (26)	587.6	246.2	0.419		
This work (prelim.)	587.6	230.6 (a)	0.392	11.92	20.29
		226.3 (b)	0.385	11.78	20.05
Average			0.402		20.09
ENDF-BV	583.5	241.2	0.413	11.92	20.43

(a) 500 $\mu g/cm^2$ $^{235}UF_4$ target

(b) 100 $\mu g/cm^2$ "

TABLE 6 COMPARISON OF THE ORIGINAL VALUES OF THE SECONDARY NORMALIZATION INTEGRALS I_1 (7.8 EV, 11 EV) AND I_2 (0.1 KEV, 1 KEV)

REFERENCE	I_1(b.eV)	I_2(b.eV)	I_2/I_1	Φ
De Saussure et al.(12)	236.7	12.30	51.97	B
Gwin et al.(25)	234.6	11.79	50.26	B
Wagemans and Deruytter(26)	240.0*	12.29 (1)	51.20	B
		12.14 (2)	50.60	
Wasson (24)	238.4*	(11.68)	(48.99)	Li
Czirr et al. (27)	244.7	11.54	47.16	Li
Muradjan et al.(28)	-	12.21*		B
Moore et al.(29)	241.2*	11.59	48.05	Li
Wagemans et al.(30)	246.2	12.54 (1)	50.93	B
		12.39 (2)	50.31	
Biriukov et al.(35)	-	12.41		
Corvi et al. (31)	241.2*	11.88	49.25	Li
This work (Prel.)	230.6 (3)	11.92	51.69	Li
	226.3 (4)	11.78	52.05	
ENDF - BV	241.2	11.92	49.42	

* Normalization value

(1) Original value with a 1/v 10B(n,α)-shape adopted

(2) Corrected for non 1/v 10B (n,α)-shape

(3) 500 μg/cm^2 235 UF$_4$ target

(4) 100 μg/cm^2 235 UF$_4$ target.

CONCLUSIONS

The present measurements yield σ_f-data in the neutron energy region from 20 meV up to 30 keV directly normalized in the thermal region. In the keV-region these data are consistent with the absolute σ_f-measurements of Szabo and Marquette (34). For the secondary normalization integral I_2 values have been obtained in agreement with those of Gwin et al. (25) and Czirr et al. (27) which were also directly normalized in the thermal region.
For the I_1 integral, however, puzzling low values have been obtained. This was also the case for $\bar{\sigma}_f$ in neutron energy intervals containing strong resonances. Three additional measurements are planned to further investigate these observations: (i) maintaining the actual ~2π-geometry but using a ^{10}B-foil for the neutron flux detection (ii) using a low detection geometry with a ^{10}B- as well as a ^{6}Li-flux monitor. Only after these measurements definite conclusions on the I_1 and I_2 integrals can be formulated and final $\bar{\sigma}_f$-values can be released.

The present study also gives some evidence for a correlation between the integral I_2 and the neutron flux monitor used. The influence of a normalization via I_1 or I_2 on the final cross-section has been shown. The magnitude of possible normalization errors is illustrated.

Finally, since ^{235}U is expected to be an "easy" nucleus (low α-activity, high σ_f-values), there are some indications that the important discrepancies still present in ^{235}U(n,f) cross-section measurements might partially be due to errors in the neutron flux determination.

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