New Integral references for neutron-induced reactions in fissile targets

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Extending the standard integrals

Historically, the principal international standards are a few constants at thermal point (the TNC table), including the four main fissile actinides: U233, U235, Pu239 and Pu241 .

It was found useful for normalization purposes to **provide integral values** in standardized energy-intervals.

First was studied the (n,f) reaction in the range 20 to 60 meV, around the thermal point.

But there are many high-resolution experiments (Tof) that start measuring at energies above a few 1eV, more easily reachable than the thermal point.

Then, new integral data on (n,f) were proposed in the RRR, with its ratios to the thermal point values.

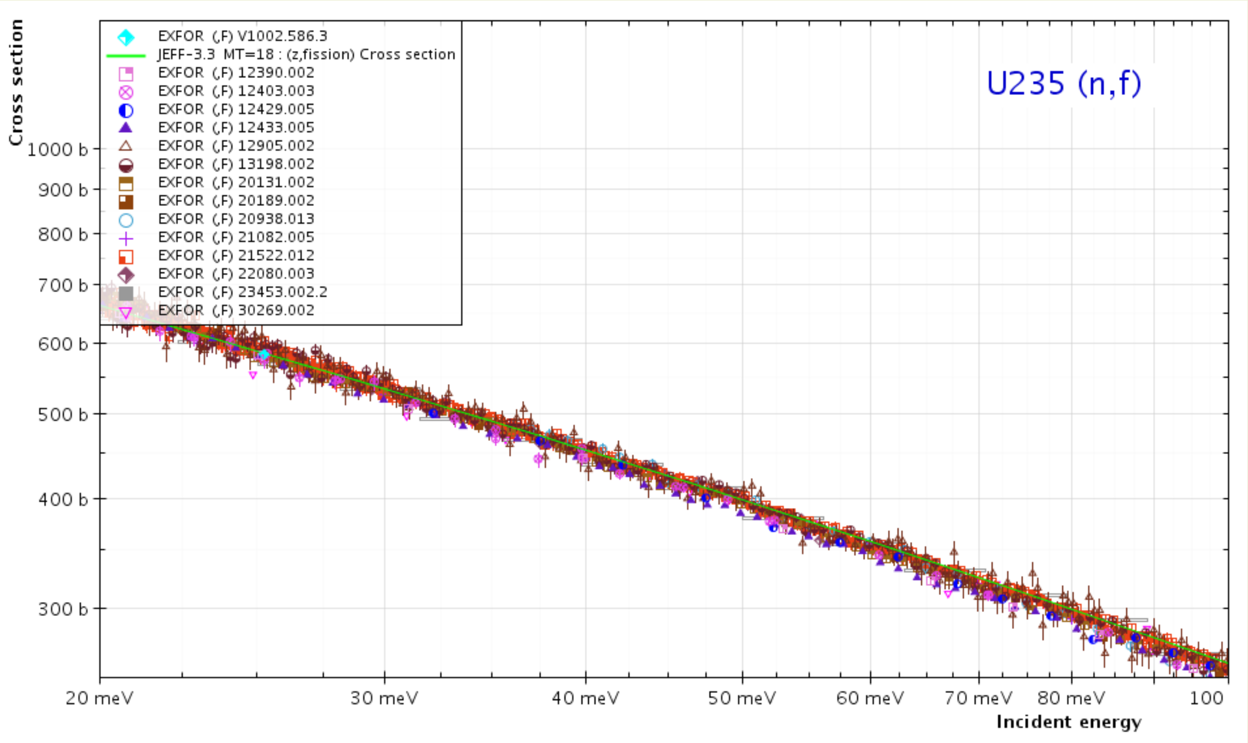
Last year, the same procedure was applied to the (n,tot) experimental data, and the corresponding (n,g) constants were deduced from the equation:

(n,g) = (n,tot) – (n,f) – (n,el) → see G.Noguere presentation

Finally, (NEW) the same procedure has been applied at energies above 1 MeV, to provide references for reactions of interest in fast-neutron reactors.

Integration interval in the thermal energy region

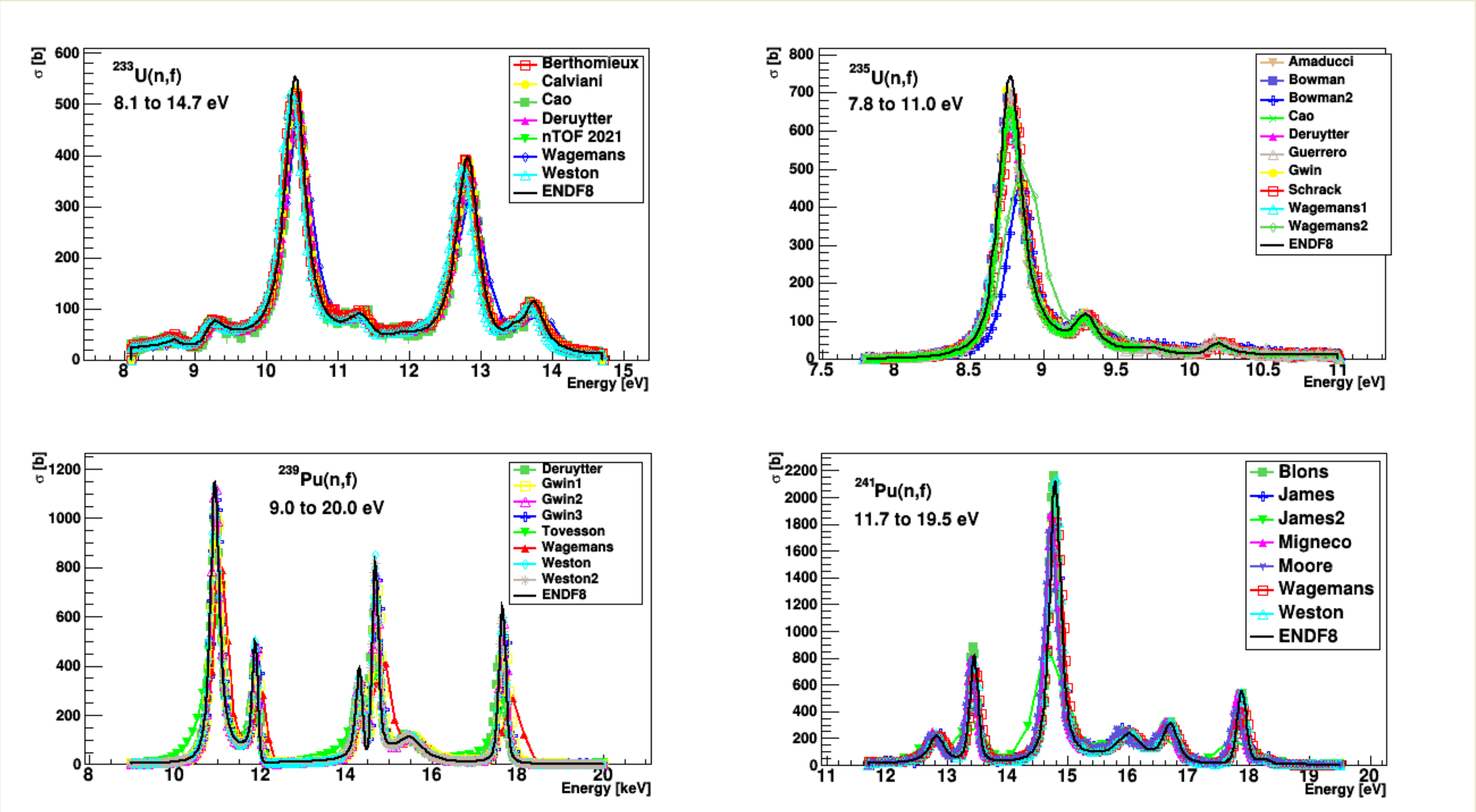
For the four fissile actinides in the TNC table experimental data were analysed by fitting it to straight-lines in log-log scale. Renormalization and energy calibration was eventually needed.



Note that the actual slope in log-log scale is not 0.5 (that correspond to the 1/v law), being different for each actinide.

Nevertheless, the straight-line approximation remains to be very accurate.

(n,f) integrals in the RRR



Here are shown the integration limits agreed to be used for the (n,f) analysis in the RRR.

The (n,f) integral for U5 has been adopted as standard in NDS2018.

Note that these same intervals were later adopted too for (n,tot) and (n,g).

First conclusions

- The values obtained by integrating in wide energy ranges are consistent, proving the usefulness of the procedure.

- Concerning (n,f), the integral values I1 and I3, and its ratios to the thermal constant, suggest the convenience of being accepted as IAEA References.

- Concerning (n,g), the high-resolution experimental datasets are not of enough good quality (mainly due to contaminants).

Therefore, the procedure has been applied to (n,tot) showing consistency of the final data, with low uncertainties, both in the thermal region and in the RRR. The (n,tot) integral values can be adopted too as References.

- Concerning (n,el), there is a lack of accurate data, pointing to the need of good experiments all along the thermal and low enegy ranges.

(n,tot) analysis: the case of U3

08/10/2022	U3(n,tot) integrals			Renormalized @ TNC18			*slope*	σ_{tot}	I1_anal
	I_therm	8.1-14.7	ratio	factor	I_therm	8.1-14.7	fit	fit	analytical
Block	19,36			1,005	19,46		-0,498	590,4	19,46
Bollinger	19,76			0,990	19,56		-0,496	593,5	19,57
Harvey1	19,28	863,5	44,8	1,006	19,40	868,7	-0,501	588,6	19,38
Harvey2	19,52	871,5	44,6	0,998	19,48	869,8	-0,496	590,7	19,48
Kolar		872,6		1,002		874,3			
Pattenden		854,8		1,020		871,9			
Moore	19,31			1,009	19,48		-0,500	591,3	19,47
Nikitin	19,07			1,016	19,38		-0,499	587,1	19,35
Pschenichnyj	19,46			1,002	19,50		-0,503	592,1	19,48
Safford1	19,45			1,005	19,55		-0,483	589,1	19,52
Safford2	19,34			1,008	19,49		-0,492	589,4	19,47
Mean value	**19,39**	**865,6**	**44,72**		**19,48**	**871,17**	**-0,496**	**590,2**	**19,46**
ENDF8	19,44	873,1	44,9		19,44	873,1	-0,493	588,5	19,43
JEFF 3.3	19,45	872,9	44,9		19,45	872,9	-0,492	588,7	19,44
JENDL4	19,45	872,9	44,9		19,45	872,9	-0,492	588,7	19,44
Mean value	**19,45**	**872,97**	**44,89**		**19,45**	**872,97**	**-0,492**	**588,6**	**19,44**

- The final results are highlighted in bold. They have as reference U5(n,f) at thermal point to be 587 b (NDS18).

- Note the agreement between I1 renormalized and I1 from the analytical fit.

These two remarks are of interest: every actinide result is related to U5(n,f); and the integral-reference procedure give us very accurate results.

(n,tot) integral results

(n,tot)	σtot | TNC | %	I1_renorm	I3_renorm	I3 / I1	σtot / I1
U233	590.2(1.3) 590.1(2.5) 0.0	19.5(0.1) 0.3%	871.2(2.5) 0.3%	44.8	30.31(17)
U235	700.7(1.3) 700.9(1.9) 0.0	22.5(0.1) 0.3%	375.0(10) 2.7%	16.7	31.16(19)
Pu239	1028.7(1.1) 1030.8(3.5) -0.2	35.2(0.1) 0.3%	1834(38) 2.1%	52.1	29.21(18)
Pu241	1392.1(2.1) 1398(13) -0.4	45.9(0.1) 0.2%	2235(94) 4.2%	48.7	30.35(06)

Note the close agreement with TNC for the thermal XS

(TNC here quoted is the sum of (n,f)+(n,g)+(n,el) in the NDS-2018 table)

Uncertainties of the integrals are given as the Std Dev of the experiments:

note the low values for I1 and the high ones for I3)

The integration procedure applied to higher energies NEW!

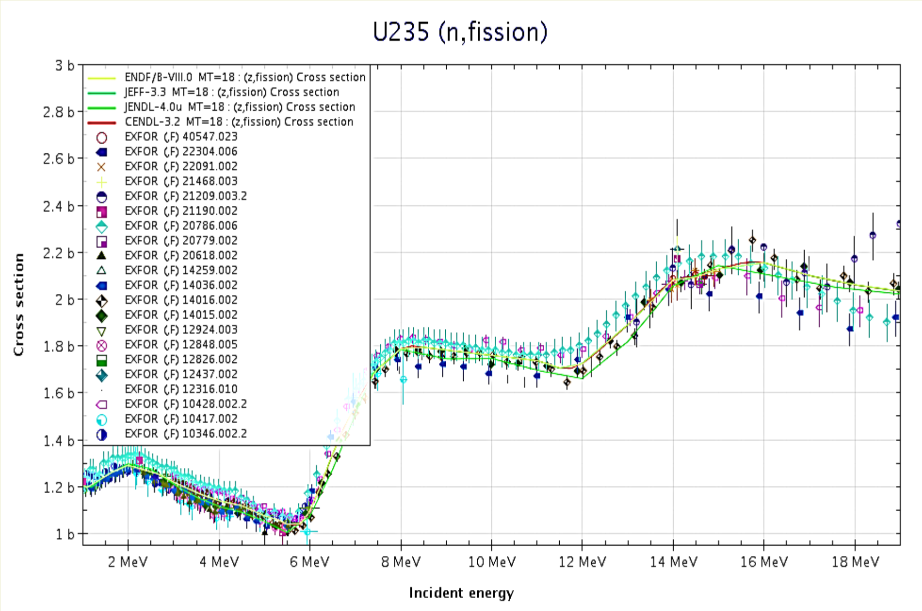
Now the question is to look for high quality references for fission above 1 MeV, in order to improve the present standards and, in consequence, the evaluation of those actinides involved in the fast-reactors cycle.

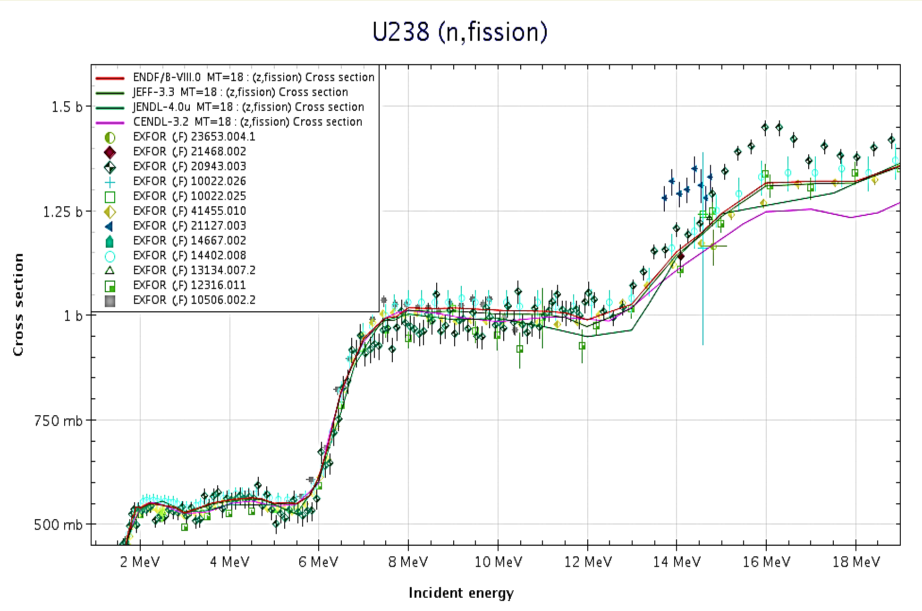
Therefore, the first step is to adopt the integration limits.

Ideally, the integration value should be high enough to minimise the statistical uncertainties, and to be very independent of eventual energy miscalibrations.

Above 1 MeV there are two plateaus fulfilling these requirements:

Two plateaus

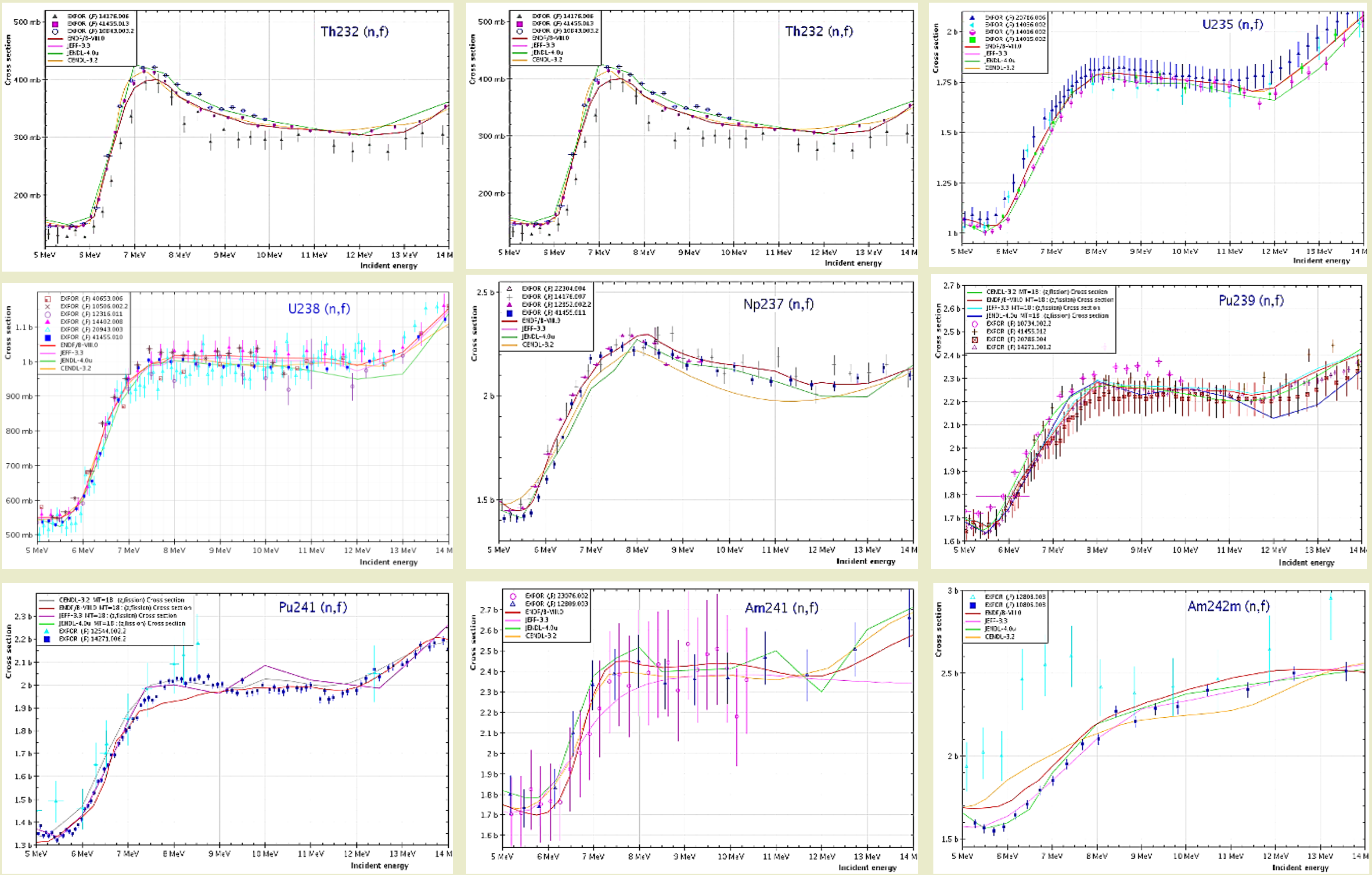




The first plateau has been more widely measured and it is always very useful for absolute measurements using the well known self-fission spectrum of Cf252 sources.

On the other hand, the second plateau is more useful for ToF experiments because its XS values are higher and flatter, without sharp changes in the FF anisotropy..

Plateau at 9 MeV



This general trend around 9MeV for the whole group of actinides makes useful to define a unique interval of integration.

In this preliminary work the interval from 8 to 10 MeV is proposed.

Measuring ratios of integrals

Once adopted the same integration interval for the whole set, their ratios become important constraints at the evaluation time.

It is worth noting that both U5(n,f) and U8(n,f) are IAEA Standards, and as a matter of fact, the best-known quantity is the ratio of both XS.

This ratio was analyzed in detail in the paper on the USU. Different statistical models were used, giving finally a value at 9 MeV of 0.572, with an uncertainty of 0.3%.

If the ratio at 9 MeV is taken from the Standards (NDS2018) is 0.571

In the recent paper on the evaluation of Cf SACS in the 1 to 5 MeV interval, a renormalization is proposed, giving a ratio of 0.573

In the present work we evaluate the mean value of the ratios at 9 MeV obtained by fitting to straight-lines the 19 datasets retrieved from EXFOR giving us a value of 0.570(2), 0.4% statistical uncertainty.

This is showing that the method of fitting to straight lines (as it was done in the thermal energy range) is reliable enough.

The new value of 0.572 can be hold as reference

Three step method

The second step after adopting the integration limits is to select those high-resolution datafiles to be used to obtain the integral reference value.

All the selected datafiles have been retrieved from EXFOR, rejecting as outliers, eventually, those showing either anomalous dispersion or an integral value not compatible with the mean value of the others.

Every experimental datafile has been, eventually, renormalized using as factor the ratio of its declared monitor/reference to the present Standards.

Finally, the points in every dataset falling in the selected interval have been fitted to straight-lines, giving so the fitted value at 9 MeV and the integral value.

The case of U8(n,f)

24/05/2023	U8(n,f) 8-10 MeV integrals					
	year	EXFOR	XS@9MeV	factor	XS Renorm.	Integral 8-10
Meadows	1975	10506 002 2	0,991	1,010	1,001	2,002
Smith	1957	12316 011	0,975	1,020	0,995	1,989
Tovesson	2014	14402 008	1,031	1,005	1,036	2,072
Leugers	1976	20943 003	0,987	1,020	1,007	2,013
Scherbakov	2002	41455 009	0,990	1,005	0,995	1,990
Pankratov	1963	40653 006	0,994	0,990	0,984	1,968
Mean value			**0,995(10)**		**1,003(9)**	**2,006(18)**
ENDF8			1,017		1,014	2,028
JEFF 3.3			1,009		1,007	2,014
CENDL 3.2			0,999		0,998	1,996
JENDL4			0,989		0,994	1,988
Mean value			**1,004(9)**		**1,003(6)**	**2,007(13)**

Integrals from ratio U8(n,f) / U5(n,f) [mean value of XS U5 = 1,768 b]					
	year	EXFOR	Mean	*(1)	*(2)
Meadows	1975	10906 002	0,577	1,020	2,040
Difilippo	1978	10635 002	0,568	1,004	2,008
Behrens	1977	10653 004	0,564	0,997	1,994
Lisowski	1991	14016 003	0,577	1,020	2,040
Tovesson	2014	14402 009	0,579	1,024	2,047
Casperson	2018	14498 002	0,562	0,994	1,987
Cierjaks	1976	20409 002	0,560	0,990	1,980
Coates	1975	20414 002	0,573	1,013	2,025
Paradela	2015	23269 002	0,573	1,013	2,026
Jie Wen	2020	32798 002	0,580	1,025	2,051
Goverdovski	1983	40831 003	0,565	0,999	1,997
Scherbakov	2002	41455 002	0,562	0,994	1,987
Mean value			**0,570(2)**	**1,008(4)**	**2,015(8)**

Six XS datafiles retrieved from EXFOR:
The fitted values at 9 MeV are renormalized;
The uncertainties have been calculated from the standard deviation to the non-weighted mean values.

The first column shows the point-values at 9 MeV in the evaluated libraries, and in the second one are the values after fitting in the same way that for the experimental datasets.

The coincidence is in prove of the straight-line hypothesis

The same procedure is applied to the 12 ratios to U5 retrieved from EXFOR.

In column (1) are the XS values obtained after multiplying by the U5 value taken as reference and in column (2) are their corresponding integrals.

Wich is the best STANDARD?

Actually, we have three values derived from different experiments (not fully uncorrelated): the XS of U5, the XS of U8 and their ratio.

Let's take as reference the U5(n,f) @ 9 MeV because there are many ratios to them of the whole set of actinides measurements.

Let's take the U5(n,f) XS @ 1 MeV = 1.766 b,
derived from the renormalized fits of the U5(n,f) experimental datasets, following the present method.

It is worth mentioning that no matter this number is, what it is important is to have a main reference (to be changed, eventually).

So, if the ratio U8/U5 is 0.572, the U8(n,f) XS @ 9 MeV becomes 1.010 b,
to be compared with the point-wise value given by GMA for the NDS of 1.017(14) b, and with the mean value 1.003 b given by both the integrals procedure and the evaluated libraries.

Table of XS at 9 MeV

By applying the same method to other actinides we have the following values of the XS @ 9 MeV

	XS [b] at 9 MeV			
	From EXFOR (XS renorm.)	From EXFOR (Ratios to U5)	Evaluated libraries Point value	Fitted value
Th232	0.339(6)	0.345(5)	0.339(4)	0.342(4)
U233	2.287(18)	2.291(24)	2.262(13)	2.259(10)
U235	1.768(17)	-----	1.771(13)	1.769(7)
U238	1.003(9)	1.008(5)	1.004(9)	1.003(6)
Np237	2.245(55)	2.234(29)	2.166(35)	2.180(32)
Pu239	2.273(21)	2.237(23)	2.253(11)	2.260(5)
Pu241	2.016(##)	1.992(14)	1.970(9)	1.994(15)
Am241	2.416(3)	2.570(264)	2.395(20)	2.397(24)
Am242m	2.329(80)	2.324(84)	2.273(28)	2.257(31)

This evaluation of the actinides out of the TNC table can be used for renormalization purposes, with reference to U5 = 1.766 b

Table of integral values

	Integral 8-10 MeV [b·eV]		
	From EXFOR	From EXFOR (Ratios to U5)	Evaluated libraries
Th232	0.678(12) 2%	0.689(9) 1.4 %	0.684(8)
U233	4.573(38) 0.8%	4.581(48) 1%	4.519(19)
U235	3.535(35) 1%	-----	3.537(13)
U238	2.006(18) 0.9%	2.016(8) 0.4%	2.007(13)
Np237	4.417(82) 2%	4.404(59) 1.4%	4.360(65)
Pu239	4.546(42) 1%	4.474(46) 1%	4.521(10)
Pu241	4.032(##)	3.983(28) 0.7%	3.988(31)
Am241	4.832(6) 0.14%	5.140(528) 10%	4.794(48)
Am242m	4.658(160) 3.5%	4.648(168) 3.4%	4.514(63)

There is a general good agreement with the mean value of the evaluated libraries.

Uncertainties are derived from the standard deviation from the mean values. Therefore, it takes the higher values where the number of datasets is low.

CONCLUSIONS

-Integral references in both the thermal energy range and in the RRR were defined, probing to be very useful for evaluators.

They are easy to be used for normalizing both old and new experimental datasets.

- Both (n,f) and (n,tot) integral references at low energies are based on consistent experimental datasets, and can be taken as Standards for both U5 and U8.

- Now is the turn to define a new integrating interval above 1 MeV in order to better normalize the evaluated datafiles, first for those in the TNC table and then for those being of interest for new nuclear technology.

An interval is proposed in the second plateau, showing that the fitting to straight-lines in the 8 to 10 MeV range provide good enough values including most of the actinides involved in the fast-neutron reactors.