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Fission Measurements. Part 2

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Outline – Part II

Status of fission data on minor actinides

- Libraries
- Previous data
- New results

The measurements at n_TOF Other methods (surrogate) Conclusions



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Data analysis



Data analysis

$$\sigma_{f} (^{A} X, E_{n}) = ratio (E_{n}) \cdot \sigma_{f} (^{235} U, E_{n})$$

Standard cross-section used as
reference (from evaluated data file)
$$ratio(E_{n}) = \frac{C_{A_{X}} - B_{A_{X}}}{C_{235_{U}} - B_{235_{U}}} \cdot \frac{N_{235_{U}} \cdot \varepsilon_{235_{U}} cf_{235_{U}}}{N_{A_{X}} \cdot \varepsilon_{A_{X}} cf_{A_{X}}}$$

Things to **remember** about the use of ²³⁵U (or ²³⁹Pu) as reference samples:

- reference samples typically mounted inside the same chamber for same efficiency
- all samples with the **same area** to avoid correction for the flux interception
- if possible, same thickness, to minimize efficiency corrections (ε)
- approximately same count-rate, to minimize dead-time correction (cf)
- need to correct for **anysotropy** in angular distribution of fission fragments (particularly important at high energy). Included in the factor cf.

Possible sources of background

Several sources of background may affect the measurements of fission cross-sections:



It is preferable to try and **minimize** all possibile sources of background, to increase signal-to-background ratio and minimize uncertainty on background subtraction:

- high neutron flux (to minimize ambient background and natural radioactivity)
- minimize mass of the detector and surrounding material (for neutron scattering)



When extracting the fission cross-sections with ratio method, **uncertainties** related to:

mass of the sample and of the referencetypically, 1 %presence of other isotopes (contaminants) in the samplesdepends on the samplebackground subtractiondepends on the samplewrap-around neutronsdepends on the facilityefficiency and dead-time correctionsdepends on detector, few %neutron beam attenuationdepends on set-up, < 1%</td>evaluated cross-sections used as referencetypically, 1-3 %

In addition, other possible sources of uncertainty are:

- sample **non-uniformity** (combined with beam non-uniformity)
- **misallignement** between sample and reference (don't intercept the same neutron flux)



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Sensitivity analysis shows that current uncertainties for MA are too high for the design of Gen IV e ADS burners

Isotope		Uncer	tainty	Isotope		Uncertainty		Isotope		Uncertainty	
Cross-	Energy Range	(?	6)	Cross-	Energy Range	(%)		Cross-	Energy Range	(%	6)
Section		Initial	Target	Section		Initial	Target	Section	~	Initial	Target
	19.6 - 6.07 MeV	29.3	20.1	Fe56 _{Ginei}	19.6 - 6.07 MeV	13.0	8.9	Am242m σ _{fiss} Pu240 σ _{fiss}	6.07 - 2.23 MeV	23.4	8.0
U238 _{Øinei}	6.07 - 2.23 MeV	19.8	4.6		6.07 - 2.23 MeV	7.2	4.1		2.23 - 1.35 MeV	19.7	8.2
	2.23 - 1.35 MeV	20.6	4.5		2.23 - 1.35 MeV	25.4	3.3		1.35 - 0.498 MeV	16.5	4.3
	1.35 - 0.498 MeV	11.6	5.5		1.35 - 0.498 MeV	16.1	3.2		498 - 183 keV	16.6	3.1
	0.07 - 2.23 MeV	14.2	6.5	Pu239 _{Teapt}	1.35 - 0.498 MeV	18.2	10.1		183 - 67.4 keV	16.6	3.1
	2.23 - 1.35 MeV	21.3	5.8		498 - 183 keV	11.6	6.5		67.4 - 24.8 keV	14.4	4.1
	1.35 - 0.498 MeV	16.6	3.4		183 - 67.4 keV	9.0	5.6		24.8 - 9.12 keV	11.8	4.3
	498 - 183 keV	13.5	2.6		67.4 - 24.8 keV	10.1	6.3		9.12 - 2.03 keV	12.4	6.5
Pu241	183 - 67.4 keV	19.9	2.6		24.8 - 9.12 keV	7.4	5.5		2.03 - 0.454 keV	12.2	5.2
σ_{fiss}	67.4 - 24.8 keV	8.7	3.3		9.12 - 2.03 keV	15.5	6.7		19.6 - 6:07 MeV	9.6	8.6
	24.8 - 9.12 keV	11.3	3.5	016	19.6 - 6.07 MeV	100.0	62.3		6.07 - 2.23 MeV	4.8	2.8
	9.12 - 2.03 keV	10.4	5.4	б _{сарt}	6.07 - 2.23 MeV	100.0	39.5		2.23 - 1.35 MeV	5.7	2.6
	2.03 - 0.454 keV	12.7	4.4	Na23	2.23 - 1.35 MeV	12.6	9.3		1.35 - 0.498 MeV	5.8	1.8
	454 - 22.6 eV	19.4	8.6	σ _{inel}	1.35 - 0.498 MeV	28.0	4.0		498 - 183 keV	3.9	3.9
U238	24.8 0.12 keV	0.4	20		6.07 - 2.23 MeV	31.3	8.2		2.03 - 0.454 keV	216	12.4
σ _{capt}	24.0 - 7.12 KeV	7.4	3.0	Cm244	2.23 - 1.35 MeV	43.8	8.2				
يرجع معرجين معرجينا	<u>, </u>		· · · · · · · · · · · · · · · · · · ·	σ _{fiss}	1.35 - 0.498 MeV	50.0	5.1				
					498 - 183 keV	36.5	12.1				

Table 4. ABTR, SFR, EFR: Uncertainty Reduction Requirements to Meet Integral Parameter Target Accuracies

Source: Aliberti, Palmiotti, Salvatores, NEMEA-4 workshop, Prague 2007

Isotope	2	Uncer	tainty	Isotope		Uncertainty		Isotope		Uncertainty	
Cross-	Energy Range	. (?	6)	Cross-	Energy Range	(%	6)	Cross-	Energy Range	(?	6)
Section		Initial	Target	Section		Initial Target		Section		Initial	Target
	19.6 - 6.07 MeV	29.3	9.0		498 - 183 keV	15.0	2.9	Pu240 offiss Si28	6.07 - 2.23 MeV	4.8	2.9
	6.07 - 2.23 MeV	19.8	2.0	B10	183 - 67.4 keV	10.0	2.7		2.23 - 1.35 MeV	5.7	2.6
U238	2.23 - 1.35 MeV	20.6	2.1	Grant	67.4 - 24,8 keV	10.0	3.3		1.35 - 0.498 MeV	5.8	1.6
σ _{inel}	1.35 - 0.498 MeV	11.6	2.3	vcapt	24.8 - 9.12 keV	8.0	3.9		498 - 183 keV	3.9	3.7
	498 - 183 keV	4.2	3.8		9.12 - 2.03 keV	8.0	6.0		2.03 - 0.454 keV	21.6	11.8
	183 - 67.4 keV	11.0	4.2		1.35 - 0.498 MeV	18.2	6.6		19.6 - 6.07 MeV	52.9	72
	6.07 - 2.23 MeV	14.2	5.0		498 - 183 keV	11.6	4.4	σ _{capt}	19.0 - 0.07 Met	52.5	·
	2.23 - 1.35 MeV	21.3	3.9	Pu239	183 - 67.4 keV	9.0	4.0	Si28	6.07 - 2.23 MeV	13.5	3.9
	1.35 - 0.498 MeV	16.6	2.1	σ _{capt}	67.4 - 24.8 keV	10.1	4.2	σ _{inel}	2.23 - 1.35 MeV	50.0	7.4
	498 - 183 keV	13.5	1.7		24.8 - 9.12 keV	7.4	3.8	Pb206 _{Ginel}	6.07 - 2.23 MeV	5.5	4.2
Pu241	183 - 67.4 keV	19.9	1.7		9.12 - 2.03 keV	15.5	3.2		2.23 - 1.35 MeV	14.2	4.0
σ _{fiss}	67.4 - 24.8 keV	8.7	1.9	016	19.6 - 6.07 MeV	100.0	37.9		1.35 - 0.498 MeV	9.2	4.7
	24.8 - 9.12 keV	11.3	2.0	σ_{capt}	6.07 - 2.23 MeV	100.0	37.9	Pb207 σinel Pb	6.07 - 2.23 MeV	5.0	4.9
	9.12 - 2.03 keV	10.4	2.1	Am243	6.07 - 2.23 MeV	17.9	4.9		2.23 - 1.35 MeV	13.8	6.0
	2.03 - 0.454 keV	12.7	2.1		2.23 - 1.35 MeV	35.3	3.9		1.35 - 0.498 MeV	11.3	3.6
	454 - 22.6 eV	19.4	5.4		1.35 - 0.498 MeV	42.2	2.3		607 - 223 MeV	54	3.0
	6.07 - 2.23 MeV	31.3	3.0	σ_{inel}	498 - 183 keV	41.0	3.7	σ_{inel}	0.07 - 2.25 WeV	5.4	5.0
C=244	2.23 - 1.35 MeV	43.8	2.6		183 - 67.4 keV	79.5	3.7	Am243	6.07 - 2.23 MeV	11.0	2.3
Cin244	1.35 - 0.498 MeV	50.0	1.5		67.4 - 24.8 keV	80.8	12.4		2.23 - 1.35 MeV	6.0	1.9
Unss	498 - 183 keV	36.5	4.0		1.35 - 0.498 MeV	23.4	21.4	Ofiss	1.35 - 0.498 MeV	9.2	1.7
	183 - 67.4 keV	47.6	7.3		498 - 183 keV	16.5	6.3	Bi209	2.23 ~ 1.35 MeV	34.1	2.8
U238	24.8 - 9.12 keV	9.4	1.8	Am242m	183 - 67.4 keV	16.6	4.7	$\sigma_{\rm inel}$	1.35 - 0.498 MeV	41.8	4.3
σ_{eapt}	9.12 - 2.03 keV	3.1	1.8	$\sigma_{\rm fiss}$	67.4 - 24.8 keV	16.6	4.8	N15	2.23 - 1.35 MeV	5.0	3.1
E-56	6.07 - 2.23 MeV	7.2	2.6		24.8 - 9.12 keV	14.4	5.6		1.35 - 0.498 MeV	5.0	1.2
reso	2.23 - 1.35 MeV	25.4	1.7		2.04 - 0.454 keV	11.8	5.9	σει	498 - 183 keV	5.0	1.9
Oinel	1.35 - 0.498 MeV	16.1	1.5	Na23	1.25 0.408 May	28.0	10.5		183 - 67.4 keV	5.0	2.3
				σinel	1.55 - 0.498 MeV	20.0	10.5	Zr90	607 222 14-11	19.0	2.2
								σ_{inel}	0.07 - 2.25 MCV	18.0	5.5

 Table 5. ABTR, SFR, EFR, GFR, LFR, ADMAB: Uncertainty Reduction Requirements to Meet

 Integral Parameter Target Accuracies

	Isotope	Cross-	8- Energy Range	Uncert. (%)		Isotope	Cross-	Energy Range	Uncert. (%)					
Isotope		Section	Energy runge	Initial Target		Locope	Section	Energy Range	Initial Target					
			6.07 - 2.23 MeV	31.3	3.0	Bi209 Am243	σ _{inel} σ _{fiss}	2.23 - 1.35 MeV	34.1	2.8				
	Cm244	σ _{fiss}	2.23 - 1.35 MeV	43.8	2.6			1.35 - 0.498 MeV	41.8	4.2				
			1.35 - 0.498 MeV	50.0	1.5			6.07 - 2.23 MeV	11.0	2.3		N	ed to ir	n
10		σ _{inel}	6.07 - 2.23 MeV	7.2	2.5			1.35 - 0.498 MeV	9.2	1.6				
	Fe56		2.23 - 1.35 MeV	25.4	1.6	Cm244	ν	6.07 - 2.23 MeV	11.1	2.5		accu	iracy to	le
			1.35 - 0.498 MeV	16.1	1,5			1.35 - 0.498 MeV	5.5	1.3		2 ($\frac{1}{2}$ on mo	אר
			1.35 - 0.498 MeV	42.2	2.3	N15	σ_{el}	1.35 - 0.498 MeV	5.0	1.2				
	Am243	$\sigma_{\rm inel}$	498 - 183 keV	41.0	3.6	Pb	σ_{inel}	6.07 - 2.23 MeV	5.4	2.9		from	om few	k
			183 - 67.4 keV	79.5	3.7	Zr90	σ_{inel}	6.07 - 2.23 MeV	18.0	3.3	i l		several	٨
		σ_{fiss}	1.35 - 0.498 MeV	16.6	2.1	Pu238	σ _{fiss}	2.23 - 1.35 MeV	33.8	6.0	ı L		Jeverar	
ł	Pu241		498 - 183 keV	13.5	1.7			1.35 - 0.498 MeV	17.1	3.4				
			183 - 67.4 keV	19.9	1.7			498 - 183 keV	17.1	3.9	l			
		σ_{fiss}	6.07 - 2.23 MeV	11.7	1.7	Cm242	σ _{fiss}	6.07 - 2.23 MeV	52.6	26				
	Am241		2.23 - 1.35 MeV	9.8	1.4			498 - 183 keV	66.0	28.4	1			
			1.35 - 0.498 MeV	8.3	1.2			1.35 - 0.498 MeV	7.0	2.8				
			1.35 - 0.498 MeV	49.4	3.3	14230		498 - 183 keV	7.0	3.4				
	Cm245	$\sigma_{\rm fiss}$	498 - 183 keV	37.2	2.9	2m242m	ι σ _{fīss}	498 - 183 keV	16.6	4.8	l			
	CIII243		183 - 67.4 keV	47.5	2.9			183 - 67.4 keV	16.6	4.8				
			67.4 - 24.8 keV	-26.5	3.2	Sourc	e: A	liberti, Palm	iotti,	Salv	ato	ores,	NEMEA-	4
						work	shop. I	Prague 2007						

Table 2. Uncertainty Reduction Requirements Needed to Meet Integral Parameter Target Accuracies

ed to improve racy to less than 6 on most MA, om few keV to several MeV

Experimental data are collected in a database (EXFOR) available on many Web sites

- data in a specific format
- various information on the measurement

Evaluated data are a combination of theoretical calculations and experimental data available. Collected in different databases, available on Web:

- ENDF (American database)
- JEFF (European)
- JENDL (Japanese)
- BRONDL (Russian)

Most important web sites for cross-section retrieval: IAEA, NEA, NNDC, Korean, etc... Various programs allow to retreive data and make plots. In the following, used the the program Janis 3.0 <u>www.nea.fr/janis</u> Ask Alberto if interested in excercises on how to retrive data.



The ²³⁷Np(n,f) cross-sections

The ²³⁷Np(n,f) reaction: evaluated data libraries



Discrepancies between major cross-section databases exist in the energy region important for fast reactors.



The ²³⁷Np(n,f) reaction: data



The ²³⁷Np(n,f) reaction: new results

Recent data from LANL and n_TOF (shown later).

²³⁷Np/²³⁵U Fission Cross Section Ratio versus Energy



Systematic uncertainties limited to 1.5% over large region. Together with n_TOF data, should resolve existing discrepancies and improve accuracy of databases. First "unique" measurement from thermal to 200 MeV.



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The 237Np(n,f) cross-sections at n_TOF



NFN

Data taken with Parallel Plate Avalanche Counter (coincidence method) Analysis still in progress



The Pu(n,f) cross-sections

The ²³⁸Pu(n,f) reaction: evaluated data libraries



The ²³⁸Pu(n,f) reaction: the data



NFN

Below threshold few data available (and with scarse energy resolution) Above 1 MeV few data available (but

ICTP - IAEA, Trieste 2008

Incident energy (MeV)

0,5

17

10

The ²⁴⁰Pu(n,f) reaction: evaluated data libraries



The ²⁴⁰Pu(n,f) reaction: the data



The ²⁴¹Pu(n,f) reaction: evaluated data libraries



The ²⁴¹Pu (n,f) reaction: the data



Data are scarse and with some discrepancies ENDF currently based only on Behrens



The ²⁴²Pu(n,f) reaction: evaluated data libraries



The ²⁴²Pu(n,f) reaction: the data



The ²³⁹⁻²⁴¹Pu (n,f) reaction: new results from LANL





The Am(n,f) cross-sections

The ²⁴¹Am(n,f) reaction: evaluated data libraries



A recent sensitivity analysis indicates that uncertainty has to be reduced at high energy:

• 500 keV – 6 MeV from 10 % to 2 %



The ²⁴¹Am(n,f) reaction: data at low energy



Need more data (high resolution and high accuracy) to check databases Different data sets in different energy regions. Data collected at n_TOF for this isotope, from thermal to 200 MeV



The ²⁴¹Am(n,f) reaction: data at low energy



Above threshold, discrepancies between data of 15 %. Current evaluation mainly based on Dabbs-83.

Need new data to solve accuracy and improve uncertainties in current databases



The ²⁴¹Am(n,f) reaction: data above threshold



The ²⁴³Am(n,f) reaction: evaluated data libraries



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The ²⁴³Am(n,f) reaction: data at low energy



Very few data below threshold, and with bad energy resolution.

Different data sets in different energy regions.

Evaluation mainly based on theoretical models, adjusted to reproduce in average measured crosssections.

At low energy, need more data (high resolution and high accuracy) Experimental difficulty: contamination of ^{242m}Am !!



The ²⁴³Am(n,f) reaction: data above threshold



The ²⁴³Am(n,f) reaction: new results





The ^{242m}Am(n,f) reaction: data



Difficult measurement, due to very short half life (t_{1/2}=141 y) At high energy, problem with one data set.

Need more data

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Evaluated databases mostly in agreement Very high cross-section at low energy (10⁴ b !!)



The Cm(n,f) cross-sections
The ²⁴³⁻²⁴⁴Cm(n,f) reaction



The ²⁴⁶⁻²⁴⁸Cm(n,f) reaction



The ²⁴⁵Cm(n,f) reaction: evaluated data libraries





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The ²⁴⁵Cm(n,f) reaction: data at low energy



The ²⁴⁵Cm(n,f) reaction: data at high energy



Difficult measurement due to the short half-life New results are coming out (n_TOF, Russia)



The n_TOF results

The n_TOF data

Need new and accurate data on neutron induced fission on MA:

•Wide **energy range** (easier to check normalization)

•High resolution for Resolved Resonance Region

•Minimize possible contribution of the **background**

•Extend measurements at higher energies (> 20 MeV)

•Include new data in the **evalution**, and consider data previously neglected (if any).

New data on fission cross-sections have been collected at the **n_TOF facility** cat CERN, taking advantage of the innovative features of neutron beam

The aim is to provide accurate cross-section data that could help fulfilling the requests.



Neutron flux at n_TOF



Clear advantages of the n_TOF facilities for measuring cross-sections of radioactive isotopes

The fission measurements at n_TOF



Measured isotopes:

•²³⁵U, ²³⁸U (standard reference)
 •²³²Th, ²³³U, ²³⁴U, ²³⁶U (Th/U cycle)
 •²³⁷Np, ^{241,243}Am, ²⁴⁵Cm (transmutation and Gen IV)

The fast ionization chamber (FIC):

Detect only one fission fragment (still efficiency around 100 %)
Stack of up to 10 samples in the beam
Does not require very thin samples





The n_TOF samples

lsotope	Total mass (mg)	Mass uncertainty (%)	Half-life	Activity (sample)
²³⁵ U	31.8 (2 samples)	1.57	7.04E8 y	0.2 kBq
²³³ U	28.8 (4 samples)	1.73	1.6E5 y	5 MBq
²⁴¹ Am	2.26 (8 samples)	1.33	432 y	76 MBq
²⁴³ Am	4.8 (8 samples)	2.0	7370	7.4 MBq
²⁴⁵ Cm	1.71 (4 samples)	1.75	8500 y	0.2 GBq

²⁴³Am is contaminated at 2.6% with ²⁴¹Am ²⁴⁵Cm is contaminated at 6.6 % with ²⁴⁴Cm ($t_{1/2}$ =18 y) SF typically 10⁻⁷-10⁻¹⁰, except for ²⁴⁴Cm for which 10⁻⁴ dN/dt=N_{at}·ln2/t_{1/2} 1 y = 3.15e7 s



Time-of-flight reconstruction



Calibration of neutron energy in ToF measurements

The neutron energy (at low energy) is determined from the time-of-flight according to:



The "time-start" (or **physical reference**) for the time-of-flight is typically given by a **"prompt flash"** (γ-rays, high-energy charged particles, etc...).

T_c=correction term

In **moderated** neutron beams, L does not coincide with the **geometric distance** from neutron source, since neutrons travel some distance inside the target/moderator.

The "moderation distance" v-t depends on the neutron energy (t=moderation time).

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Necessary to calibratate the neutron energy with respect to energy standards

Calibration of neutron energy in ToF measurements



The pulse height distribution



Notes: the ²³⁵U is virtually background-free, and the amplitude distribution is as expected from simulations of the energy deposited in the gas.

All other isotopes show a large α -background (in some cases pile-up of α -particles)



Background determination and subtraction



Threshold on amplitude is the primary method for α -background and electronic noise rejection. However, for some isotopes, a large background is still present above threshold, and HAS TO BE SUBTRACTED



The ²³⁵U(n,f) reactions



First reaction to be analysed.

Gives an idea of the background and energy resolution.

NOTE: complete energy range in ONE measurement only

$$C(E_n) = \sigma_f(E_n) \cdot \Phi(E_n) \cdot N_{235} \cup \varepsilon \cdot cf$$





The ²³³U(n,f) reaction



If accuracies of few percent is required, small effects (like differences in efficiency due to different target thickness), may affect the uncertainty.

Such effects can only be estimated with very realistic Monte Carlo simulations.



The ²³³U(n,f) reaction at n_TOF



reactor technology (U/Th fuel cycle).

Fission cross-sections measured for the first time **simultaneously** from thermal energy to 500 MeV. Reached **accuracy** ~ 3 %

Risolved resonance region extended up to 10 keV



F. Belloni et al., Nucl. Sci. Eng., in preparazione



The ²³³U(n,f) reaction: the RRR





In general, at low energy, current databases seem to work pretty well.

Few details need to be considered, such as energy and strength of individual resonances.



The ²³³U(n,f) reaction: the RRR



The ²³³U(n,f) reaction: the URR



The ²³³U(n,f) reaction: group cross-sections



At thermal energy, agreement between data and evaluated libraries within 1 % !! At high energy, evaluated cross-section are underestimated by up to 12 % !! Typical example of importance of accurate new data in the whole energy region.

I N F N

The ²⁴⁵Cm(n,f) reaction at n_TOF



The results of ²⁴⁵Cm(n,f) at low energy



Comparison with previous data



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Comparison with previous data (2)

²⁴⁵Cm (n,f) cross section, 300 bin/decade



Comparison with previous data (3)





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The ²⁴⁵Cm(n,f) reaction: high energy



The ²⁴¹Am(n,f) reaction at n_TOF



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The ²⁴³Am(n,f) reaction at n_TOF



The ²³⁸U(n,f)/²³⁵U(n,f) cross-section ratio



The ²³⁸U(n,f)/²³⁵U(n,f) ratio

(sub) threshold region



The surrogate method





Fission cross-se	ections that can be possibly studied with surrogate reactions:
²³⁷ Np(n,f)	²³⁸ U(³ He,t) ²³⁸ Np
²³⁶ U(n,f)	²³⁸ U(³ He,α) ²³⁷ U
²⁴² Cm(n,f)	²⁴³ Am(³ He,t) ²⁴³ Cm
²⁴³ Cm(n,f)	²⁴³ Am(³ He,d) ²⁴⁴ Cm
²⁴⁴ Cm(n,f)	²⁴³ Am(³ He,p) ²⁴⁵ Cm

Main problem associated with the angular momentum of the CN (in surrogate reactions may be substantially different from the neutron-induced reaction).

Several model-dependent corrections have to be applied.

In some cases, the only possibility to estimate fission cross-sections


There is strong need of **accurate new data** on neutron-induced fission cross-section of several isotopes involved in **advanced nuclear reactors**.

Since a few years, renewed efforts on the experimental side, with **improved facilities** and **experimental techniques**.

Important contribution comes from the n_TOF facility at CERN: ²³²Th, ²³³U, ²³⁴U, ²³⁵U, ²³⁶U, ²³⁸U, ²³⁷Np, ²⁴¹Am, ²⁴³Am and ²⁴⁵Cm

Energy range from **thermal** to several **MeV** (in some cases up to 500 MeV)

Preliminary results indicate that current accuracy **can be improved**, at least for some isotopes (lower **background**, consistent **normalization** in the whole energy range, better **energy resolution**, etc...)

Few percent accuracy may be reachable for some isotopes.

Measuring all what is needed will take efforts and contribution from many facilities around the world, **BUT IT IS NECESSARY**.



Thank you for your attention