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ABSOLUTE MEASUREMENT OF THE PU-239 FISSION CROSS-SECTION AT 4.8, 8.65 AND 18.8 MEV NEUTRON ENERGY USING THE TCAPM

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1. INTRODUCTION

The ^{239}Pu fast neutron fission cross-section is of great importance for the predetermination of important parameters of fast breeding reactors because of the high ^{239}Pu inventory. That's why an improvement of the cross-section data accuracy from the reached value (5-10%) to the demanded (2-5%) in the 3-20 MeV energy range is a problem of first priority in nuclear data resource /1/.
In addition to cross-section measurements relative to the ^{235}U standard, precise absolute measurements seem to be ingeniously for getting additional possibilities of normalization.
Within the bounds of the joint fission cross-section measurement program of the Khlopin Radium Institute (KRI) Leningrad and the Technical University of Dresden (TUD), absolute measurements of ^{239}Pu fast neutron fission cross-section were performed in the 1983-85 years, using the time-correlated associated particle method (TCAPM) for three different energy points. The measurements were carried out at the 5 MV tandem van-de-Graaff accelerator of the CINR Rossendorf, GDR, and resulted in preliminary values of (1.740 ± 0.035)b, (2.350 ± 0.044)b and (2.487 ± 0.088)b for neutron energies of (4.8 ± 0.20) MeV, (8.65 ± 0.20) MeV and (18.8 ± 0.20) MeV, respectively.

2. EXPERIMENTAL PROCEDURE

2.1. Measuring principle

The principle of our TCAPM fission cross-section measurements (Fig.1) was already described in detail in earlier publications /2,3,4,5/. Therefore only the most important facts and parameters are to be summarized.
An associated particle (AP) detector coupled with a fast AP detection system counts charged associated particles, which are produced in the $D(d,n)^3He$ or $T(d,n)^4He$ reaction correlated with the simultaneously generated neutrons. The AP aperture A determines the cone of counted AP and selects thereby the cone of correlated

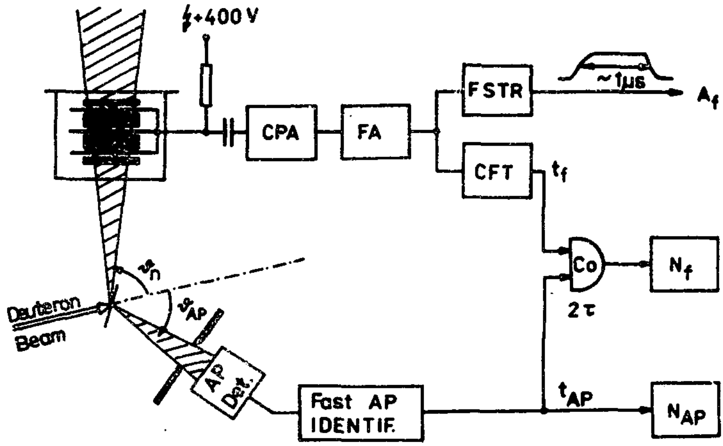


Fig.1: Measuring principle of the KRI/TUD fission cross-section measurements

neutrons, which is completely intercepted by the homogeneous fission foils placed inside an ionization fission chamber. Neutron cone profile and position are checked continuously during the runs. For each of the detected fissions and each of the detected AP the fast timing signals t_f and t_{AP} are generated, respectively. By counting the t_{AP} pulses (N_{AP}) and the coincidence signals N_f between t_{AP} and t_f pulses, the cross-section σ_f can be determined from the formula (1), if the number of fissionable nuclei per unit of area (n) in the fission foils is known:

$$\sigma_f = N_f \,/\, (N_{AP} \cdot n) \qquad (1)$$

Those of the necessary corrections which depend on the actual experimental conditions are determined from the following pulse hight spectra which are collected during the whole measuring time by means of a CAMAC system /5,6/:

- the t_{AP}-t_f time-of-flight spectrum for the correction of random coincidences within the t_{AP}-t_f coincidence window;
- the fission chamber spectrum gated by the N_f coincidence signal

for the correction of the counting losses caused by the CFT threshold in the fast t_f channel;

- AP amplitude spectrum for the background correction of the AP-channel.

2.2. Neutron producing systems

For the absolute measurements energy points were selected, where the fission cross-section shows a relatively small energy dependence within the limits of the energy distribution of the neutrons which are correlated to the registered AP.
Because of the necessary deuteron energies in the MeV region,

Neutron energy			
- Averaged energy $\bar{E}_n$ (MeV)	4.8 ± 0.2	8.65 ± 0.2	18.8 ± 0.2
- Half width ΔE_n (MeV)	0.25	0.45	0.60
Neutron production reaction	$D(d,n)^3He$	$D(d,n)^3He$	$T(d,n)^4He$
Deuteron beam			
- Energy (MeV)	5.80	9.50	6.00
- Collimator	2 apertures ø 3 mm		
- Current (nA)	400-600	400-600	500-800
Neutron producing target			
- Foil	deuterated polyethylene		self supporting Ti-T covered by 0.2mg/cm^{2}Al
- Thickness	(0.6-0.8) mg/cm^2		3.6 mg/cm^2
- Diameter	ø 35 mm		ø 16 mm
- Inventory	> 95% $(CD_2)_n$		1.3 Ci/cm^2
- Angle relative to the beam axis	52°	75°	40°
- Rotation	$(1-3)s^{-1}$	$(1-3)s^{-1}$	$(1-3)s^{-1}$
Neutron cone profile			
- Measured FWHM	4.5°	5°	6°
- Measured FW 0.005M	11.5°	12.5°	14°
- Calculated total width	10°	11°	12.5°

Tab.1: Parameters of the neutron producing systems (neutron cone profile was measured using a plastic scintillator outside the vacuum chamber (angular extent ~ 0.7°))

selfsupported targets had to be used for the neutron production. A rotating target holder system allowed to work at deuteron beam currents of $\gtrsim$500 nA continuosly $\sim$1-2 days if a deuterated polyethylene target is used, or without noticeable escape of T out of the foil if a Ti-T target is used. Kinematical and geometrical conditions as well as target properties are carefully optimized for each neutron energy point to achieve a maximum of accuracy of the neutron detection based on the AP counting.

Tab.1 gives a summary of important parameters. Neutron cone profile and energy distribution of the in-cone neutrons were calculated considering the true experimental geometry, including the finite beam focus and the target foil thickness.

The shifts of the averaged neutron energies in comparisation with our ^{235}U *fission cross-section measurements* /5,7,8/ are caused by small changes of the experimental conditions (deuteron energy and/or AP detection angle).

2.3. Associated particle detection system

The AP detection system is based on a Si(SB) detector telescope, which was already described in ref. /5,7/. The AP identification is performed

- by single channel analysis of a particle-significant spectrum containing all events within the window of the total energy spectrum (at 4.8 and 8.65 MeV neutron energy); or
- by single channel analysis of the total energy spectrum containing only events, which are identified as alpha particles within the particle-significant spectrum (at 18.8 MeV).

The determination of the background portion at the 8.65 MeV measurement is similar to the 4.8 MeV measurement. It is based on measured background spectra and described in /5,7/. The larger uncertainty of the AP background correction for the presented 18.8 MeV measurement compared with the result in /5/ is caused by the higher beam current, which leads to a rate of scattered deuterons $\sim 3\cdot10^{5}s^{-1}$ and therefore to a decreased energy resolution in the AP identification channel. Typical spectra at the 8.65 MeV measurement are shown in Fig.2; fundamental features of the AP detection systems are listed in Tab.2.

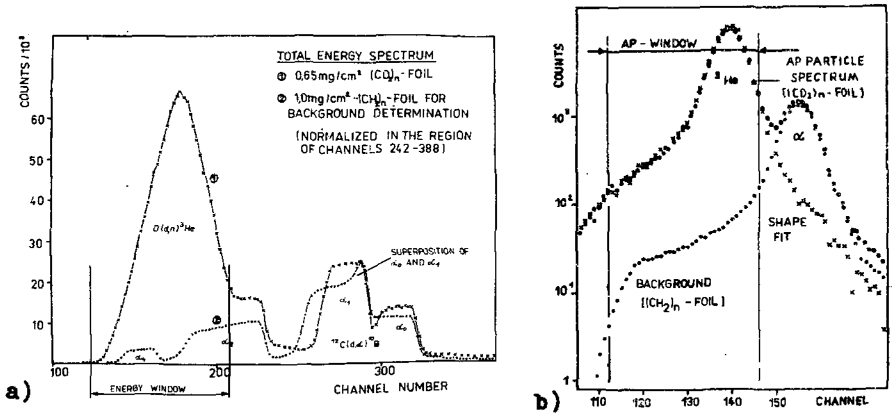


Fig.2: Total energy spectrum (2a) and particle-significant spectrum (2b) at the 8.65 MeV cross-section measurement

	Cross-section measurement at neutron energy (MeV)		
	4.8	8.65	18.8
AP detection angle ϑ_{AP}	36,5° ± 1°	42.5° ± 1°	68° ± 1°
AP cone aperture	∅5mm; a=118mm	∅5mm; a=118mm	∅9mm; a=89mm
Si(SB) detector telescope			
- Detector thickness: ΔE	11μm	9μm	10μm
E_r	52μm	48μm/52μm	72μm
- LAR output pulse length	0.5μs	0.5μs	0.25μs
- Energy resolution (^{241}Am):			
E_r (total alpha energy)	41keV	46keV/41keV	60keV
ΔE (energy straggling)	160keV	140keV	180keV
$\Delta E + E_r$ (calibrated)	70keV	75keV	110keV
- Time resolution $t_{\Delta E}-t_{E_r}$	2ns	1.5ns	2ns
Scattered deuteron rates	$1-1.5\cdot10^5 s^{-1}$	$6-8\cdot10^4 s^{-1}$	$2-3\cdot10^5 s^{-1}$
Max. AP counting rate	$4800 s^{-1}$	$4500 s^{-1}$	$4500 s^{-1}$
Mean AP counting rate	$3600 s^{-1}$	$3700 s^{-1}$	$3500 s^{-1}$
Mean AP background	1-3%	1-3%	4-6%
Mean uncertainty of background portion	0.2-0.6%	0.2-0.6%	1-2%

Tab.2: Main features of the AP detection systems

2.4. Fission fragment detection and optimization of the fission chamber design

The requirements which the fission chamber has to meet in absolute fission cross-section measurements, are discussed in /8,9/. Measuring the ^{239}Pu fission cross-section special problems arise from the high alpha activity of this nuclide, which limits the efficiency of fission fragment detection.

To dicriminate pile-ups of alpha pulses from fission events, a CFT threshold in the fast t_f channel is used, and therefore an approximation of the fission fragment spectrum from the threshold to pulse hight zero has to be performed to determine the fission fragment counting losses. Usually, this procedure is carried out by a linear extrapolation of the "plateau" region between alpha *pulses and the "low energy edge" of the fission fragment pulse* hight distribution. To realise this correction with the required accuracy, a sufficient broad plateau region of the fission fragment spectrum not overlayed by pile-ups from alpha pulses is necessary. This requires a carefully optimization of the fission chamber channel including the chamber design.

ELECTRONICS

The parallel plate fission chamber is connected with a fast current sensitive preamplifier (CPA). Its output pulse length is essentially determined by the drift time of the electrons in the active gas volume of the fission chamber. Therefore a minimum pulse length is achieved by using methane (high electron drift velocity of 0.01 cm/ns /10/) as chamber gas, working at a voltage of 400 V and a gas pressure of $\sim$110 kPa. The short current pulses are fed into a timing filter amplifier (FA). No differentiation is applied. In dependence on the intensity of the natural alpha fission foil activity, an integration time constant $\leq$10 ns gives an optimum compromise between energy and time information, both derived from the amplified current signal /11/ (see Fig.1). The time information is obtained by a constant fraction trigger (CFT), the energy information from the current pulse peak value using a fast stretcher (FSTR) with output pulse hight limitation /12,13/.

This method includes:

i) The effective pulse length which determines the pile-up probability is minimized. It is essentially determined only by the short current pulse length and always not enlarged by pulse forming circuits.

ii) The use of the peak current as a measure of the energy loss of the fission fragments excludes the induction effect, and the energy information becomes independent on the fragment flight direction.

iii) Because of the pulse shaping nearly without integration, the energy resolution only achieves values of 2-3 MeV (FWHM), but its influence on the plateau region in the fission chamber spectrum is smaller compared with the influence of the enlarged alpha pile-up probability due to the integration time constant in the case of charge sensitive preamplifiers.

CHAMBER DESIGN

The shape of the fission chamber spectrum is strongly influenced by the fission chamber geometry. For its optimization concerning a high counting efficiency and a good separation of the fission fragments from alpha pulses, the following points must be taken into consideration: A small distance between fission foils and electrodes is advantageously

- to obtain short current pulses and therefore low pile-up probabilities
- considering the fact, that the stopping power of fission fragments in contrary to alpha particles decreases along the trajectory in the active chamber volume
- to get a compact chamber design, which is necessary to guarantee that all deposits intercept the neutron cone completely.

On the other hand, the decrease of the distance is limited at a minimum value. At lower distances the influence of the electronic pulse shaping ($\sim$10 ns) on the CPA output pulse length is increasing. From calculations discussed in /9,14/ the following conclusions can be derived:

- If the distance d between fission foils and electrodes is small compared to the diameter of the active chamber volume and alpha particles can loose only a part of their energy within the chamber gas, the alpha pulses with high amplitudes are caused by multiple pile-ups of pulses from alpha particles, which have lost a considerable part of their energy by passing the chamber gas nearly parallel to the target surface.

- The low energy edge of the fission fragment spectrum is generated by fission fragments with emission angles perpendiculary to the deposit. At smaller distances d this edge is shifted to lower energies.
- The error contribution due to the correction of fission fragment pulses below the CFT threshold is influenced not only by the absolute value of this correction, but also its uncertainty, which depends strongly on the length of the "visible" plateau region of the fission chamber spectrum. This error contribution has a minimum value at an optimum fission foil-electrode distance d_0 depending on the diameter of the active chamber volume and the used fission foils and on the gas pressure: At distances $d > d_0$ the effect of increasing alpha pile-up probability dominates, leading to a higher absolute value of correction; at distances $d < d_0$ the diminished visible plateau region due to the shifted low energy edge of the fission fragment spectrum dominates, leading to a higher uncertainty of the correction.
- The fission foil thickness is an important parameter, which determines not only the amount of fission fragments which cannot leave the active layer, but also the plateau height and therefore the amount of pulses below the CFT threshold. It can be verified by analytical calculations, that the plateau height is proportional to the foil thickness.

The optimum d_0 value was estimated under the condition of fission foils with an active area of 21 mm in diameter and thickness $\lesssim 300\ \mu g/cm^2$, a methane pressure of ~110 kPa, and a total alpha activity of the fissile deposits ~1 MBq. A distance of 3 mm was choosed because of the required compact chamber design, although the optimum value should be larger. The maximum possible alpha activity was estimated to be ~10 MBq in the case of the chamber design characterized by the specified parameters.

In our measurements we used a fission chamber (shown in Fig.3) with a total ^{239}Pu inventory of ~4 mg (alpha activity~ 9.1 MBq). The chamber design and the used electronic equipement allow a counting efficiency > 96% for fission events in TCAPM measurements, if the neutron flux is $\gtrsim 10^3 s^{-1}$. An example for the reached quality of the fission fragment alpha particle separation is shown in Fig.4.

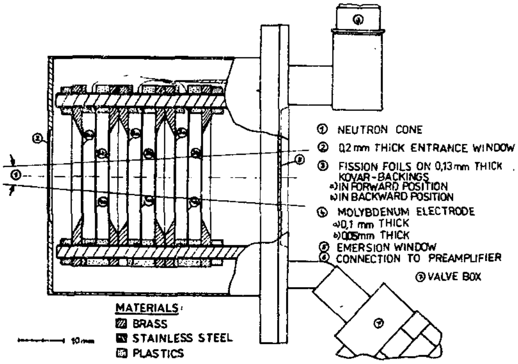
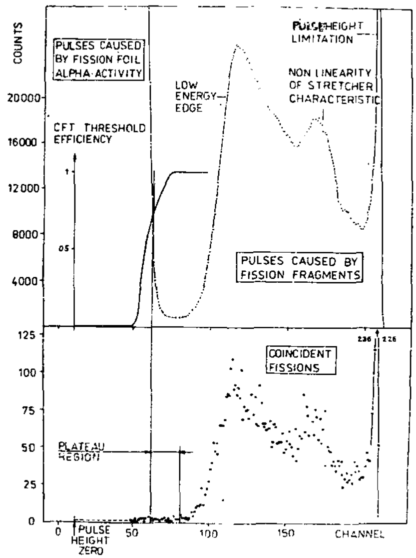


Fig.3:- Design of the used ^{239}Pu fission chamber

Fig.4: Fission chamber spectrum collected in the 8.65 MeV cross-section measurement Febr.85; measuring time: 82 hours



ADJUSTMENT OF THE CFT THRESHOLD

In our measurements the correction for fission fragment pulses below the CFT threshold is calculated from the coincident fission chamber spectrum. This spectrum contains only such alpha events, which result from random coincidences with the registered associated particles. The lower limit of the spectrum is given by the CFT threshold. One has to consider, that the CFT threshold has to be low enough to observe the full plateau region, but high enough to reduce the random coincidence rate, caused by alpha events, to a tolerable value. Although the corresponding portion N_{rc}^{α} of random coincidences within the selected window of the t_f-t_{AP} time-of-flight spectrum is included in the correction of random coincidences (calculated from the time-of flight spectrum /5/), it shold not exeed 0.5% of the coincident fission rate $\dot{N}_f$. Using the expression

$$\dot{N}_{rc}^{\alpha} = 2\tau \cdot \dot{N}_{\alpha} \cdot \dot{N}_{AP} \qquad (2)$$

(2τ - coincidence resolving time; $\dot{N}_{\alpha}$ - rate of t_f pulses due to alpha events; $\dot{N}_{AP}$ - AP counting rate), an upper limit for the $\dot{N}_{\alpha}$ rate can be derived (1):

$$\dot{N}_{\alpha} = (1/2\tau) \cdot \sigma_f \cdot n \cdot 0.5\% \qquad (3)$$

Supposing a total areal density of the fissile material $n \sim 1\ mg/cm^2$, a fission cross-section $\sigma_f \sim 2b$ and the typical value $2\tau \sim 10$ ns, a maximum value $\dot{N}_{\alpha} \sim 2\ s^{-1}$ is concluded. This consideration was used to adjust the CFT threshold before starting the experiments.

In the 18.8 MeV measurement a background portion, caused by the $^{12}C(n,n')3\alpha$ reaction at the chamber gas and correlated with the counted associated particles, had to be considered in the analysis of the fission chamber spectrum: Supposing a linear plateau shape, an additional component of events with low pulse heights was found, which cannot be explained by random alpha-AP coincidences. Therefore, a lower limit k_o was introduced which characterizes the lower end of the linear plateau region of the coincident fission chamber spectrum. The amount ε_k of correlated events below k_o was subtracted from the background-corrected number of coincident fissions. It was calculated by subtracting the amount N_{rc}^{α} of random alpha-AP coincidences, determined from the direct fission chamber spectrum and the CFT efficiency curve by means of expression (2), from the total number of events with pulse heigths $< k_o$. The true number of fission fragment pulses below k_o then was determined by linear extrapolation of the plateau region from pulse height k_0 to zero.

3. FISSION FOILS

The used fission foils (Tab.3) were prepared and calibrated at the KRI Leningrad. They were produced by thermo-sputtering using material of high isotopic purity > 99.99%. Measuring the areal density by means of low geometry alpha counting, accuracies of 1% were reached using $T_{1/2} = (2.411 \pm 0.003) \cdot 10^4$ a /15/. A portion of alpha activity from $^{238}Pu \lesssim 2\%$ had to be considered.

Pos.	Areal density ($\mu g/cm^2$)	Angular extent at the measurements 4.8 MeV	8.65 MeV	18.8 MeV
1/F	287.2 x)	25.2°	26.3°	22.8°
2/B	161.9 x)	22.4°	23.3°	20.6°
3/F	195.9	20.9°	21.6°	19.2°
4/B	162.7 x)	18.9°	19.5°	17.4°
5/F	198.2	17.8°	18.4°	16.6°
6/B	139.3 x)	16.4°	16.8°	15.4°

Tab.3: Parameters of the used fission foils. The angular extent results from the diameter of the fissile layer and its distance from the neutron source. F-forward, B-backward geometry; x) fission foil also used in the test measurement 1983

The measurements of the nonuniformity were carried out at the TUD corresponding to the experimental conditions of our cross-section measurement, especially the neutron cone profile. The alpha activity of selected target areas (Fig.5) was measured with an aperture of ⌀ 6 mm. The statistical uncertainty of the measurements was < 0.04%. Based on the results of this measurements for the several targets, the nonuniformity of the complete set was determined considering their true position within the fission chamber. The given value for the whole nonuniformity was calculated by quadratic addition of the standard deviation of the points outside the sample centre (0.24%) and the averaged difference relative to the central point (0.85%).

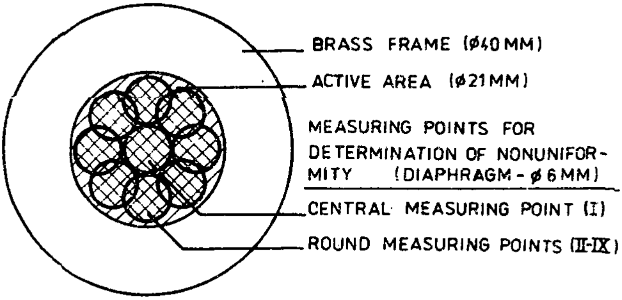


Fig.5: Fission foil nonuniformity measurement

Absolute areal density (μg/cm²)	Relative areal density difference of the outer points compared with the central point (%)							
	II	III	IV	V	VI	VII	VIII	IX
287.2	-0.17	-0.03	0.20	0.03	-0.19	0.08	-0.08	0.08
161.9	1.53	2.03	1.54	1.37	1.32	1.41	1.69	0.92
195.9	1.55	1.71	1.93	1.94	1.28	1.31	1.08	1.32
162.7	0.69	0.36	0.39	0.17	0.57	0.89	-1.05	-0.93
198.2	1.12	1.26	1.43	0.65	0.88	0.77	0.62	0.75
139.3	1.46	1.93	2.74	1.67	1.63	1.17	1.42	1.31
Summary	0.91	1.08	1.23	0.87	0.79	0.85	0.53	0.53

Tab.4: Result of fission foil nonuniformity measurements and summary for the true target position within the fission chamber used in the cross-section measurements

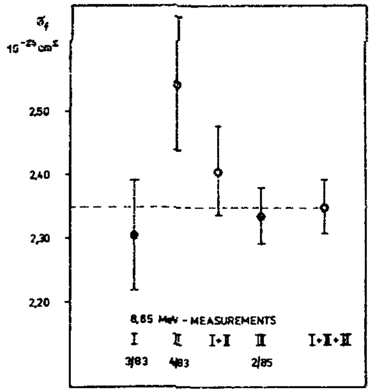
4. MEASUREMENTS AND RESULTS

In 1983 first ^{239}Pu measurements at 8.65 MeV neutron energy were performed, using a four-plate fission chamber with a total areal density of 0.7511 mg/cm^2 ^{239}Pu. The reached statistical uncertainty was ~3.5% /16,17/. Using the described fission chamber, measurements at 8.65 MeV and 4.8 MeV were carried out in Febr.85 and a short test measurement at 18.8 MeV in Nov. 1984 /18/.
The correction procedure for the cross-section calculation was described in /5/. Correction values, error contributions and

the preliminary results of the presented measurements are given in Tab.5; Fig.6 shows a comparision of the three separate 8.65 MeV measurements; Fig.7 presents our results compared with data files. To calculate the fission event losses caused by the absoption in the fission foils, an averaged fragment range of (7.5 ± 2) mg/cm^2 was assumed.

	Cross-section measurements					
	4.8 MeV		8.65 MeV (summarized)		18.8 MeV	
	Corr. (%)	Error contr. (%)	Corr. (%)	Error contr. (%)	Corr. (%)	Error contr. (%)
Counting coincidences						
-Statistics of effect	-	1.27	-	1.08	-	2.52
-Random coincidences	0.64	0.11	1.86	0.17	4.55	0.63
Fission chamber efficiency						
-Correlated background	-	-	-	-	0.34	0.13
-Extrapolation to zero	1.50	0.31	1.04	0.24	2.57	0.85
-Fragment absorption	1.21	0.46	1.20	0.43	1.30	0.39
AP counting						
-Background	2.30	0.36	1.62	0.32	5.92	1.74
Neutron cone						
-Neutron scattering	0.25	0.40	0.36	0.40	0.34	0.40
-Effective fission foil thickness due to the cone aperture	0.08	0.05	0.07	0.05	0.12	0.08
-Cone neutrons outside the angular extent of the fission foils	not corrected					
Fissile layers						
-Areal density	-	1.00	-	1.00	-	1.00
-Inhomogenity	-	0.88	-	0.85	-	0.88
Preliminary result σ_f(b)	1.740±0.035		2.350±0.044		2.487 ± 0.088	
$\Delta\sigma_f/\sigma_f$	2.00%		1.85%		3.55%	

<u>Tab.5:</u> Corrections, error contributions and preliminary results of the presented Pu-239 fission cross-section measurements



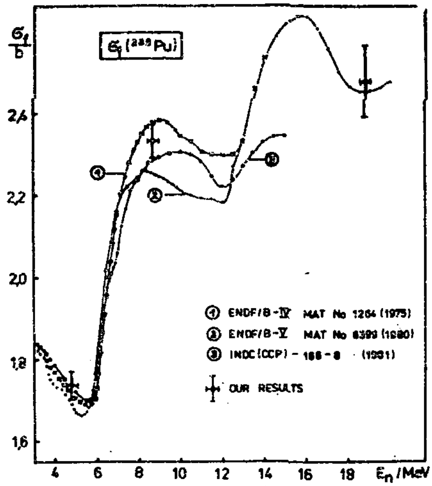


Fig.6: Results of the separate 8.65 MeV measurements

Fig.7: Presented results compared with data files

Further efforts will be directed to more precise measurements of the areal density of the fission foils, to the analysis of the neutron cone profile outside the angular extent of the fission foils caused by charged particle scattering within the neutron producing target and to experimental investigations for the determination of the fission chamber efficiency.

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