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Measurement of neutron induced fission of ^{235}U, ^{233}U and ^{245}Cm with the FIC detector at the CERN n_TOF facility

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Abstract. A series of measurements of neutron induced fission cross section of various TRU isotopes have been performed at the CERN n_TOF spallation neutron facility, in the energy range from thermal to nearly 250 MeV. The experimental apparatus consists in a fast ionization chamber (FIC), used as a fission fragment detector with a high efficiency. Good discrimination between alphas and fission fragments can be obtained with a simple amplitude threshold. In order to allow the monitoring of the neutron beam and to extract the n_TOF neutron flux, the well known cross section of the ^{235}U(n,f) reaction, considered as a fission standard, has been used. Preliminary results for the cross section are shown for some selected isotopes such as ^{235}U, ^{233}U and ^{245}Cm in the energy range from 0.050 eV to about 2 MeV.

1 Introduction

A major problem in the development of nuclear energy production is the issue of nuclear waste treatment and storage. The most significant radiotoxicity contribution comes from various transuranic elements (TRU) such as Np, Pu, Am and Cm that are build up as a result of multiple neutron captures and radioactive decays in the presently operating nuclear reactors based on the U/Pu nuclear fuel cycle. A solution could come from incineration or trasmutation, via neutron induced fission of TRU's, in subcritical systems, such as an Accelerator Driven System (ADS), or in critical systems, such

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as future Gen-IV (fast) nuclear reactors. Some isotopes, e.g., the fissile ^{233}U, are important for lighter fuel cycles, such as the Thorium Fuel Cycle, interesting for the lower production of minor actinides and breeding possibilities. More precise and complete basic nuclear data are required for the engineering design and safety evaluation of such devices [1].

Since the current data for the involved isotopes are scarce and contradictory (especially for Cm and Am isotopes), the n_TOF Collaboration has performed measurements of neutron induced fission cross sections of ^{233}U, ^{245}Cm, ^{241,243}Am and of the fission standards ^{235}U and ^{238}U at CERN n_TOF facility [2].

2 The n_TOF facility

The n_TOF facility is a time of flight installation based on a spallation neutron source. Neutrons are produced by a sharply pulsed, 6 ns wide, 20 GeV/c proton beam impinging into a $80 \times 80 \times 60\,\text{cm}^3$ lead block surrounded by 6 cm of H_2O that works as moderator and coolant. The protons are provided by the CERN Proton Synchrotron (PS) which can operate in dedicated ($7 \cdot 10^{12}$ protons per pulse) or in parasitic mode ($4 \cdot 10^{12}$ protons per pulse). The main features of this facility are the wide neutron energy range (from thermal to 250 MeV) high instantaneous flux ($3 \cdot 10^5$ n/cm^2/pulse at the measuring station with the 8 cm diameter collimator for the fission setup), low duty cycle (averaged 0.25 cm^{-1}), high energy resolution in the Experimental Area ($1.1 \cdot 10^{-3}$ at 30 keV) and low background. A measuring station is located at the end of the n_TOF tunnel, 187.5 m from the spallation target and delimited by two walls 7.5 m apart. Several polyethylene absorbing blocks as well as iron and concrete are used in order to reduce the overall background.

3 Experimental setup

The detection setup for the measurement of the neutron induced fission cross section is based on a Fast Ionization Chamber (FIC) used as a fission fragment detector. It is constituted by a stack of several parallel-plate chambers with 5 mm spacing between electrodes and operating with argontetrafluormethane (90% Ar + 10% CF_4) at 720 mbar pressure. The whole detector is an assembly of 17 ionization chambers, with a total of 16 targets and 18 electrodes, mounted together in a chamber of 50 cm length, thus allowing a simultaneous measurement of the fission cross sections of several isotopes. Each target consists of a stainless steel holder and a 100 μm thick aluminum foil backing, with very thin layers of target material (4 – 450 μg/cm^2) on both sides. A circular area of 80 mm diameter was deposited on the backing using the painting technique [4]. Two detectors have been used in the n_TOF measurements: FIC-0 was essentially dedicated to low activity isotopes, whereas FIC-1 was used for highly radioactive species like ^{233}U, ^{241,243}Am and ^{245}Cm, and is qualified as a "sealed source" compliant with the ISO 2919 norm. No gas circulation is needed, since the ionization chamber is not working in proportional mode.

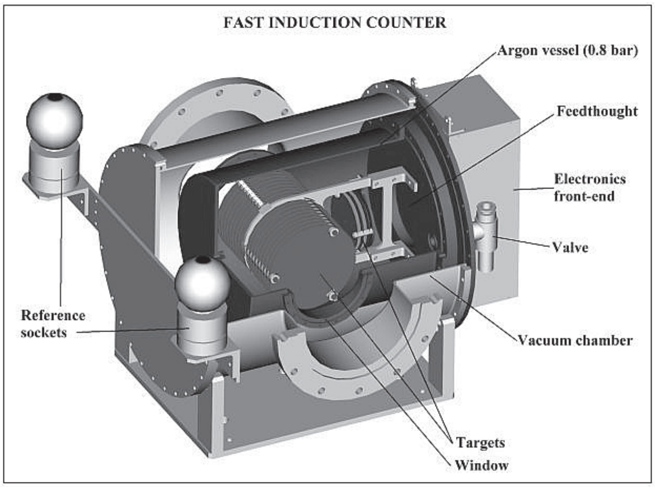

Fig. 1. Experimental setup constituted by a series of 17 parallel plate ionization chambers, including 16 targets. The chambers are filled with argontetrafluormethane at 720 mbar pressure. The magnitude of the electric field between the gaps is 600 V/cm.

Table 1. Masses and uncertainties for the various TRU samples.

Sample	Mass (mg)	Error (mg)
^{235}U	31.8	0.5
^{233}U	28.8	0.5
^{245}Cm	1.71	0.03
^{241}Am	2.26	0.03
^{243}Am	4.8	0.1

Since some of the isotopes are characterized by an activity that reaches tens of MBq, the characteristics of the n_TOF beam (i.e., the high instantaneous flux and low repetition rate) enables us to measure the fission cross section of those nuclei with high accuracy for the first time. The detector signals were acquired with a set of Acqiris Flash Analog to Digital Converters (FADC) modules, sampling the full waveform of the detector signal (at 100 MS/s) and recording the digitized detector pulses for each event or neutron bunch, thus allowing the complete information from fission events to be recorded for repeated and detailed off-line analysis. It is worth mentioning that, since the n_TOF neutron flux is determined from the ^{235}U(n,f) reaction, all others measured cross section are extracted relatively to the fission cross section of ^{235}U. Since this is considered a fission standard, a good accuracy (in the order of ≃5% due mainly to mass uncertainties and statistics) is expected for the other measured samples.

The total masses of the samples are reported in table 1. These values along with the various impurities have been obtained by alpha spectrometry using a silicon detector.

4 Results

The required informations needed for data analysis are: the signal amplitude, total charge, time of flight and the baseline value, that are determined for each pulse signal. Additionally,

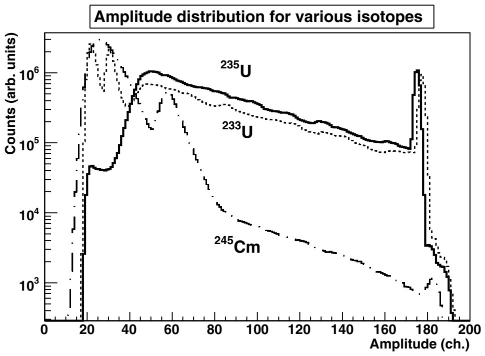

Fig. 2. Amplitude distributions of the signals for the various isotopes. The peaks at channel ≃180 are artifacts due to saturation of the FADC signals.

for each proton bunch, the beam intensity and the peak of the initial flash corresponding to ultrarelativistic muons travelling at a velocity very close to speed of light, are extracted: this last information is used as a reference for the extraction of the absolute neutron time of flight. In figure 2 the amplitude distributions for the various isotopes are presented: it is clear that, in the case of ^{235}U and ^{233}U, a simple amplitude threshold works well for the discrimination between fission fragments and α particles; for the ^{245}Cm sample, the presence of a large pileup between α particle complicates the analysis, since a higher threshold has to be applied, which cuts also a large fraction of fission fragments. A subsequent normalization of the cross section has to be performed. For the time-to-energy calibration, in order to consider the "effective" neutron flight path (sum of the geometrical length and of the "moderator distance"), a method developed in [5] has been taken into account. Additionally ^{235}U(n,f) resolved resonances were used for the energy calibration procedure. An example of the results for a limited energy interval is reported figure 3. A very good agreement with known experimental data and with ENDF/B-VII.0 is observed.

4.1 Neutron flux

For the determination of the absolute value of the cross sections of the actinide targets, the ^{235}U isotope was used for the extraction of the n_TOF neutron flux in the fission configuration (that is with the 8 cm diameter collimator, see [3]). The energy behaviour of the neutron flux can be derived from the ratio between the ^{235}U fission rate and an evaluated cross section. The results are shown in figure 4. It is important to underline that the histogram represents the product between that value and the efficiency of the detector, which is however close to 100% for the applied threshold.

The new evaluated library ENDF/B-VII.0 has been chosen as the reference library. As predicted by the simulations and according to the physics of the spallation source, the flux peaks at thermal energy (due to the water moderation effects), is approximately isolethargic between 0.25 eV and 10 keV and then peaks again around 1 MeV due to evaporation from the spallation products. Some clear dips due to absorption in the aluminum alloy windows at the end of the spallation targets

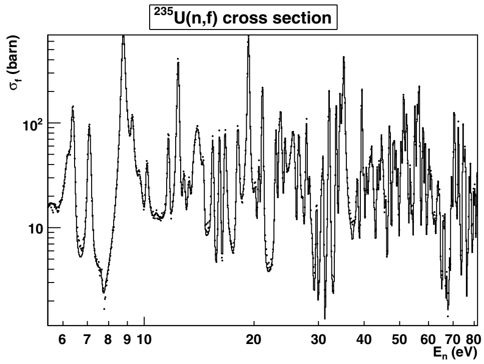

Fig. 3. ^{235}U(n,f) cross section (dots) in the interval between ≃ 6 eV and 60 eV, compared to ENDF/B-VII.0 (continuous line).

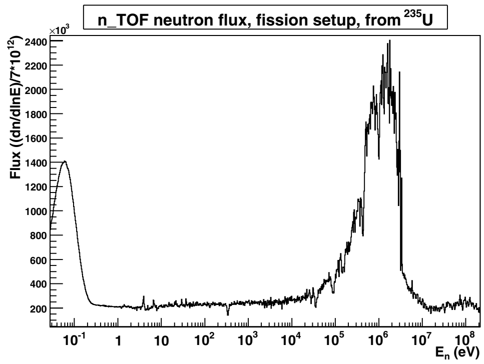

Fig. 4. Representation of the neutron flux for the fission mode in lethargy scale. This flux is obtained as the ratio between the one based on ENDF/B-VII.0 for ^{235}U. The two small peaks in the flux at 3.3 eV and 6.6 eV are probably due to some incorrect evaluation of two dips in the ENDF/B-VII.0.

and due to the presence of oxygen in the moderator are present. MCNPX simulations have been performed in order to quantify the beam attenuation in various materials (targets, backings and electrodes). This value has been evaluated to be at most of 0.1%, so no corrections need to be applied. The absence of large fluctuations in the resonance region clearly shows that a good reproduction of the shape of the resonances has been obtained, while above that threshold (located at ≃1 keV) fluctuations increase due to the lack of description of the resonances in ENDF/B-VII.0.

4.2 ^{233}U(n,f)

The experimental results for the ^{233}U(n,f) cross section are shown in figure 5, in comparison with ENDF/B-VII.0. It is clear from figure 2 that a higher value of the threshold with respect to the value set for ^{235}U needs to be applied; the subsequent loss of efficiency was corrected recalculating the corresponding flux from ^{235}U for the new used threshold.

Small differences in the amplitude of some ^{233}U resonances are observed, while in the resonance dips the experimental cross section appers to be lower than the evaluated data, indicating a good rejection of the background with respect to the fission fragment signals.

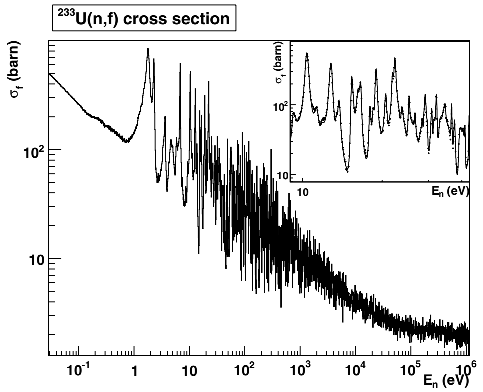

Fig. 5. ^{233}U(n,f) cross section from thermal energies up to 1 MeV. The cross section values are obtained with the flux of figure 4. The insert refers to the n_TOF data (dots) in comparison with ENDF/B-VII.0 (line) in the energy interval between 10 eV and 40 eV.

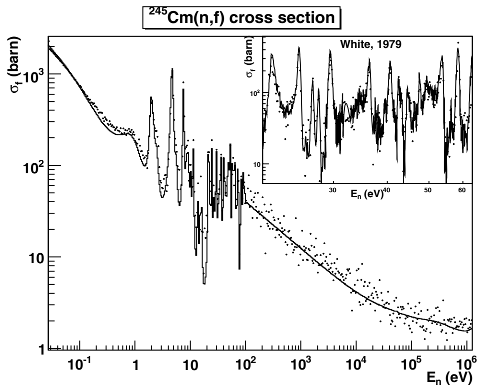

Fig. 6. ^{245}Cm(n,f) experimental cross section (dots) compared to ENDF/B-VII.0 evaluated data in the energy range from 0.035 eV to ≃1 MeV. In the insert n_TOF data (dots) are compared to White [10] data (line) in the energy interval between 20 eV and 60 eV. The n_TOF data has been normalized to previously available experimental data at thermal energy [7–9].

4.3 ^{245}Cm(n,f)

^{245}Cm is the most interesting isotope since only few and discordant experimental measurement exists; in particular no experimental results exist in the EXFOR database for the energy range between thermal and 20 eV. This isotope presents a very large α background, as evident from figure 2 (due to the relatively short halflife of 8500 y). For this reason, in addition to a higher amplitude threshold, the experimental data have been corrected also for the residual alpha background. The amount to be subtracted was determined from measurements performed without the neutron beam and with the same amplitude threshold. Due to an uncertainty in the detection efficiency, normalization to a known cross section value is required. Since at thermal energy various measurement [7–9] have been performed (although with discrepancies on the order of 10%), we have chosen a weighted mean of those values for normalization purposes.

n_TOF preliminary data, that fills the region between thermal and 20 eV where previous experimental results are not available (fig. 6), are higher than ENDF/B-VII.0, particularly in the energy range between 0.1 eV and 8 eV (where the discrepancies rise up to 10%) and in the resonances high energy shoulders. A more refined analysis is however needed before drawing a definite conclusion.

When compared with previous [10] experimental results (see insert in fig. 6), good reproduction of the fission resonances is obtained, with discrepancies up to 20% for dips and amplitudes of some resonances. Additionally, FIC data confirms a previous experimental results that shows that the ENDF/B-VII.0 doublet at $E_n \simeq 15$ eV appears as a single resonance.

5 Conclusions

Neutron induced fission cross section measurements of ^{235}U, ^{233}U, ^{245}Cm and ^{241,243}Am have been performed at the n_TOF spallation facility at CERN, using a Fast Ionization Chamber (FIC). First preliminary results for ^{235}U, ^{233}U and ^{245}Cm show results consistent with databases in the resonance region, with no normalization required for ^{233}U. In the case of ^{245}Cm, for the energy range between thermal and 20 eV, we obtained the first experimental data ever published, while showing a good agreement with previous data in the region above that value. The determination of resonance parameters is now in progress with the SAMMY code. In the near future we expect also to extend FIC measurements for actinides up to 200 MeV.

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