Study of Neutron-Induced Fission Cross Sections of U, Am, and Cm at n_TOF

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Abstract. Neutron induced fission cross sections of several isotopes have been measured at the CERN n_TOF spallation neutron facility. Between them some measurements involve isotopes (^{233}U, ^{241}Am, ^{243}Am, ^{245}Cm) relevant for applications to nuclear technologies. The n_TOF facility delivers neutrons with high instantaneous flux and in a wide energy range, from thermal up to 250 MeV. The experimental apparatus consists of an ionization chamber that discriminates fission fragments and α particles coming from natural radioactivity of the samples. All the measurements were performed referring to the standard cross section of ^{235}U.

Keywords: Neutron cross section, neutron-induced fission reactions, n_TOF
PACS: 25.85.-w,25.85.Ec,28.20.-v

INTRODUCTION

Progress in the field of Nuclear Technology requires a strong effort in order to improve the current knowledge of cross sections. Among them a better accuracy on fission neutron-induced cross sections is mandatory. The design of systems based on the Th/U fuel cycle [1], Accelerator Driven Systems (ADS) [2–4], and Gen-IV nuclear reactors [5] depends on a better determination of these cross sections.

In particular, it would be possible to increase the efficiency of the fuel cycle, improving the fuel burn-up, and to upgrade the safety of future systems. Moreover, the presence of an higher fraction of actinides in the fuel mix would be possible.

One has also to consider that nowadays one of the main limitation of nuclear energy programs is the nuclear waste treatment and storage. A significant fraction of the high-level nuclear wastes is constituted by minor actinides, and between them Am and Cm isotopes. A possible solution to the problem of nuclear waste management could be the transmutation, via neutron induced fission of transuranic elements, both in subcritical and critical systems.

In order to improve the reliability of evaluated databases [6] the n_TOF collaboration has performed several measurements of neutron induced cross sections (capture and fission). In this contribution some results on ^{233}U, ^{241,243}Am and ^{245}Cm are reported.

In particular, the ^{233}U(n,f) cross section is crucial for the study of the Th/U fuel cycle, which interest resides in the abundance of the ^{232}Th seed and in the reduced production of long-lived actinides. Moreover, discrepancies up to 15% between data from different measurements [7-10] are present in the fast neutron energy region for the ^{243}Am(n,f) cross section; stringent requirements for this cross section were found for Advanced Minor Actinides Burners (ADMAB), where a reduction of a factor 5 of the uncertainty is requested. Stronger improvements on accuracy are requested for the ^{243}Am(n,f) and ^{245}Cm(n,f) cross sections, where a reduction factor of 7 and 15, respectively, were demanded [11].

The measurements performed at the CERN n_TOF facility allowed to collect high accuracy data from thermal energy up to tenths of MeV, with this full energy range covered simultaneously, thanks to the favorable conditions offered by the neutron facility.

EXPERIMENTAL SET-UP

The measurements were performed at CERN taking advantage of the neutron beam available at the n_TOF facility [12, 13]. The unprecedented high instantaneous neutron flux in combination with the low duty cycle, high resolution, and low background of the n_TOF neutron beam allows one to collect capture cross section data with good accuracy and with an excellent signal-to-background ratio. The pulsed neutron beam of the n_TOF facility is generated in spallation reactions in a massive lead target by 20 GeV protons [13]. The spallation neutrons are slowed down and moderated in the lead target and in a 5.8 cm thick layer of cooling water surrounding the target. The resulting neutron spectrum runs from thermal energies to 250 MeV. The neutron beam is transported through an evacuated flight path with collimators at 135 and 175 m to the measuring station at a distance of 185 m from the spallation target. The beamline extends 12 m beyond the experimental area to minimize the effect of back-scattered neutrons. Background due to fast charged particles is suppressed by a 1.5 T sweeping magnet, heavy concrete walls, and a 3.5 m thick iron shielding.

Measurements of neutron-induced fission cross sections have been carried out using Fast Ionization Chambers (FIC) [14] built in collaboration between the Joint Institute of Nuclear Research (JINR, Dubna, Russian Federation), the Institute of Physics and Power Engineering (IPPE, Obninsk, Russian Federation), the Istituto Nazionale di Fisica Nucleare, and CERN. The detector consists of a stack of ionization chambers, assembled along the direction of the neutron beam, thus allowing the simultaneous measurement on several isotopes. Each chamber consists of three electrodes: a central one, 100 μm thick Al foil plated on both sides with sample material, and two 15 μm thick Al anode foils at a distance of 5 mm from the cathode, used to define the electric field. The electrodes are 12 cm in diameter, while the diameter of the sample deposit is 8 cm, so to match the size of the neutron beam. The detector is operated with a gas mixture of 90% Ar and 10% CF4 at 720 mbar pressure.

The total mass of the measured samples is reported in Table 1, together with the sample activities. The samples were prepared by means of the painting technique [15].

All detected signals were recorded with fast digitizers; a sampling rate of 100 MSamples/s was chosen, which allow to extend the time-of-flight (TOF) range to 80 ms, corresponding to a minimum neutron energy of about 30 meV. Then, the data were processed and stored by the standard n_TOF data acquisition system [16].

TABLE 1. Mass and activity of the samples involved in the measurements.

Sample	Mass (mg)	Activity
235U	31.8	0.2 kBq
233U	28.8	5 MBq
241Am	2.26	76 MBq
243Am	4.80	7.4 MBq
245Cm	1.71	0.2 GBq

DATA ANALYSIS

Cross sections were extracted referring to ^{235}U, which is an accepted standard between 0.15 MeV and 200 MeV [6]. In particular, the neutron-induced fission cross sections are extracted by Eq. 1,

$$\sigma_x(n, f) = \sigma_{235}(n, f) \cdot \frac{N_x}{N_{235}} \cdot \frac{m_{235}}{m_x} \cdot \frac{A_x}{A_{235}} \quad (1)$$

where

– σ_{235} (n,f) corresponds to the tabulated ENDF/B-VII.0 [6] cross section,
– x stands for the investigated isotope, i.e. ^{233}U, ^{241}Am, ^{243}Am or ^{245}Cm,
– N_x is the number of fission events detected for isotope *x*,
– m_x is the mass (in grams) of isotope *x*,
– A_x is the atomic number of isotope *x*

Since the investigated isotopes and the ^{235}U reference samples are mounted in the same detector, they are exposed to the same neutron flux.

The number of detected fission events obtained by Eq. 1 had to be corrected for self absorption and dead-time. The number of particles leaving the sample as well as their energy spectrum is determined by the sample thickness. Moreover, since the samples have slightly different thicknesses corresponding efficiency corrections were determined by means of detailed Monte Carlo simulations of the energy loss in the sample and in the gas volume. It was assumed that the fission events were uniformly distributed inside the sample and that the fragments were emitted isotropically. The FLUKA code was used for the simulations [17]. The loss of counts due to the dead-time induced by the reconstruction routine was calculated under the hypothesis of a non-paralyzable model. The efficiency and dead-time corrections were found to be of the order of a few percent, with a corresponding uncertainty of less than 1%.

It is important to note that the data are not normalized to any previous result, as in some past measurements, but rely only on the standard ^{235}U(n,f) cross section. For the first time, the whole energy range from thermal to tenths of MeV is covered in a single measurement, thus minimizing possible systematic uncertainties related, for example, to the absolute normalization of the cross section

RESULTS

Accurate and high resolution neutron-induced cross sections were extracted. These results will contribute to improve the reliability of cross sections databases. Detailed information on the results can be found in the referenced cited in [18]. In the following few examples of the impact of present results will be given.

Figure 1 shows how the high resolution of the n_TOF data would allow to extend the limit of resonances listed in current databases; this is particularly important to estimate self-shielding effects in reactor simulations.

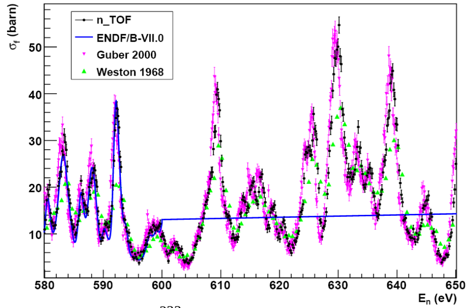

FIGURE 1. The results of the ^{233}U(n,f) reaction around 600 eV [6, 19, 20].

Figure 2 shows a comparison between present data and previous evaluations in different energy ranges [6, 19] for the ^{233}U(n,f) cross section. This figure shows a general agreement with past measurements, with the relevant exception of the energy range between 100 eV and 10 keV where databases need to be revisited.

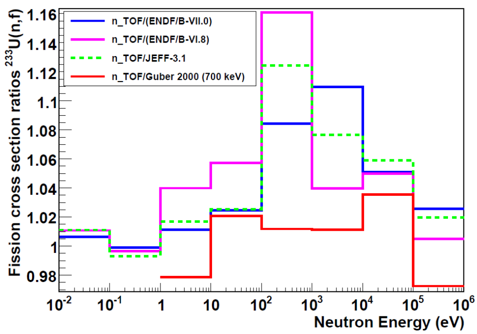

FIGURE 2. ^{233}U: ratio between n_TOF results and previous data and evaluations over neutron energy decades [6, 19].

Another important example concerns the ^{243}Am(n,f) cross section. Present data allow solving a long-standing discrepancy of more than 15%; as shown in Figure 3 n_TOF data confirm current evaluations, against recent previous results [8].

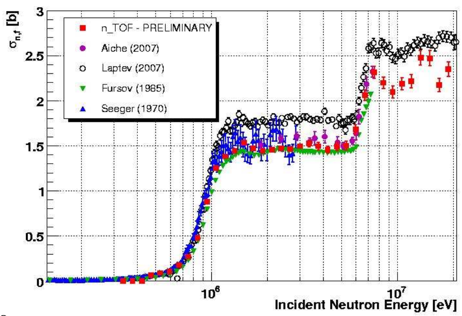

FIGURE 3. ^{243}Am:comparison between present results and previous data [7-10].

Measurements of ^{241}Am and ^{245}Cm fission cross sections were difficult because of the high α activity of the samples; this situation was also complicated by the presence of contaminants in the samples. In these cases it was possible to extract cross sections with larger uncertainties (≈10%) [18].

CONCLUSIONS AND PERSPECTIVES

Taking advantage of the high instantaneous flux and the high energy resolution of the CERN n_TOF facility, neutron-induced fission cross sections of ^{233}U, ^{241}Am, ^{243}Am and ^{245}Cm have been measured. The results are important to resolve discrepancies in previous data, thus providing a reliable basis for future evaluations.

Long-term plans of interest for fission measurements at n_TOF also include the construction of a short, 20 m flight path that would allow the study of very low mass samples and of very low cross sections, taking advantage of the unique characteristics of the n_TOF neutron beam.

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