

## Fission Cross-section Measurements of $^{233}\text{U}$ , $^{245}\text{Cm}$ and $^{241,243}\text{Am}$ at CERN n\_TOF Facility

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Neutron-induced fission cross-sections of minor actinides have been measured using the n\_TOF white neutron source at CERN, Geneva, as part of a large experimental program aiming at collecting new data relevant for nuclear astrophysics and for the design of advanced reactor systems. The measurements at n\_TOF take advantage of the innovative features of the n\_TOF facility, namely the wide energy range, high instantaneous neutron flux and good energy resolution. Final results on the fission cross-section of  $^{233}\text{U}$ ,  $^{245}\text{Cm}$  and  $^{243}\text{Am}$  from thermal to 20 MeV are here reported, together with preliminary results for  $^{241}\text{Am}$ . The measurement have been performed with a dedicated Fast Ionization Chamber (FIC), a fission fragment detector with a very high efficiency, relative to the very well known cross-section of  $^{235}\text{U}$ , measured simultaneously with the same detector.

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## I. INTRODUCTION

Development of nuclear energy programs around the world are presently closely related to the issue of nuclear waste treatment and storage. This is due to the fact that a significant fraction of the high-level waste is constituted by Pu and minor actinides, in particular Np, Am and Cm, built up as a result of multiple neutron captures and radioactive decays in current nuclear reactors. Transmutation in critical, such as Gen-IV, or subcritical (ADS) systems could be a solution; however, in order to reduce calculation uncertainties in the design and operation of new generation reactors, high precision data on neutron induced fission cross-sections from thermal neutron energies up to several tens of MeV are required for a variety of transuranic elements [1]. In particular a pressing need exists for Cm and Am isotopes, for which the available data are scarce and show large discrepancies. To address these needs the n\_TOF Collaboration has performed measurement of neutron-induced fission cross-sections of  $^{233}\text{U}$ ,  $^{245}\text{Cm}$  and  $^{241,243}\text{Am}$  which allowed collecting data from thermal up to several tens of MeV neutron energy, with the full range covered simultaneously.

## II. THE EXPERIMENTAL SETUP

The n\_TOF neutron beam is produced by spallation of 20 GeV/c protons from the CERN PS machine, which impinges on a lead target. The target is cooled with a 5.8 cm thick water layer which acts also as moderator. A detailed description can be found in Ref. 2. n\_TOF

is characterized by a very high instantaneous neutron flux ( $\sim 10^5$  n/cm<sup>2</sup>/pulse @ 200 m), which improved the ratio signals over noise, as well as by a high resolution in neutron energy, which allows a better description of the fission cross-section in terms of resonance parameters.

The detection system used for the measurements is based on a Fast Ionization Chamber (FIC), specifically built for n\_TOF. The detector and its performances are described in detail in Ref. 3. It consists in a stack of ionization chambers assembled along the direction of the neutron beam; each of them is constituted by three electrodes 12 cm in diameter, while the diameter of the sample deposit is 8 cm, so to match the neutron beam size. The detector is operated with 90% Ar + 10% CF<sub>4</sub> at 720 mbar pressure. The standard n\_TOF DAQ, based on 8 bit flash ADCs was used for these measurements [4].

A detailed off-line analysis of the digitized signals, based on ROOT routines and described in Ref. 5, allowed the extraction of relevant information of the neutron time-of-flight and on the energy deposited by the fission fragment in the detector. The total mass, together with uncertainties and sample activities, are reported in Ref. 5. The samples were prepared by means of the painting technique [6].

## III. RESULTS OF INTEREST FOR THE TH/U FUEL CYCLE - $^{233}\text{U}(\text{n},\text{f})$

New data on neutron-induced fission cross-section on  $^{233}\text{U}$ , of interest for the Th/U fuel cycle, have been already published [7] and the most important results are reported here. For all measured isotopes, the cross-section has been determined relative to the  $^{235}\text{U}(\text{n},\text{f})$  reaction. An efficiency correction, due to the different

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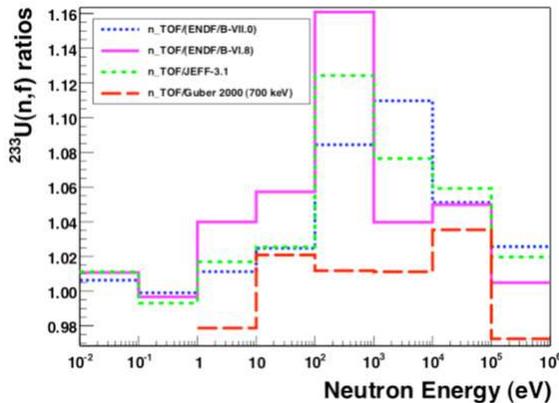


Fig. 1. (Color online) Ratio between n\_TOF results and with previous data and evaluations averaged over neutron energy decades.

thicknesses of the  $^{233}\text{U}$  and  $^{235}\text{U}$  samples have been calculated with the FLUKA code and applied to the data.

A general view of the differences between n\_TOF results and previous data and evaluations is shown in Fig. 1. While the average difference is within 2% below 100 eV, large discrepancies exist in the energy range between 100 eV and 10 keV, with the evaluations well below the data. The n\_TOF data, together with results from Guber *et al.* [8], strongly suggest a revision of the evaluations, at least above 100 eV.

The region from 1 MeV up to 20 MeV has been analyzed by a different technique, due to the presence of an intense prompt-flash [9]. The results, shown in Fig. 2, show a better agreement with the ENDF/B-VI.8 library rather than ENDF/B-VII.0, which has been adjusted to match the results of integral measurements.

#### IV. RESULTS ON ACTINIDES : $^{245}\text{CM}$ , $^{241,243}\text{AM}$

The analysis of the  $^{245}\text{Cm}(n,f)$  measurement is complicated by the presence of a significant  $\alpha$ -particle background and by the spontaneous fission contribution of  $^{244}\text{Cm}$ , present in the sample as a contamination. Both effects are corrected using the runs without beam. Nevertheless, to maximize the signal-to-background ratio, it was necessary to apply a high threshold on the pulse height distribution. This, however, results in large uncertainties in the efficiency correction. For this reason, a different approach, based on renormalization of the present data at thermal energy, has been followed in order to extract the cross-section.

Since the  $^{245}\text{Cm}(n_{th},f)$  cross-sections in the databases show a discrepancy up to 20%, we have chosen to normalize the n\_TOF data to the average of two recent results [10,11]. The final cross-section in two different energy regions is shown in Figs. 3 and 4.

As evident in Fig. 3, the  $^{245}\text{Cm}(n,f)$  n\_TOF data confirms the non- $1/v$  behavior of the cross-section below 0.1 eV, with a shape very similar to White *et al.* [12] and

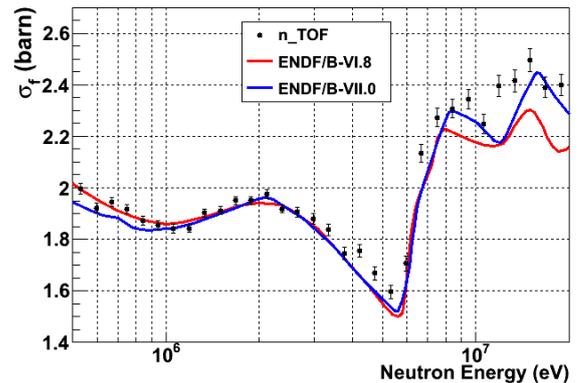


Fig. 2. (Color online) n\_TOF data compared with ENDF/B-VI.8 and ENDF/B-VII.0 between 500 keV and 20 MeV.

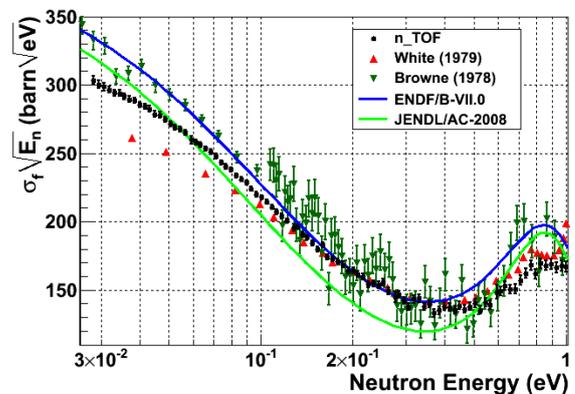


Fig. 3. (Color online)  $^{245}\text{Cm}(n,f)$  n\_TOF data between 0.03 and 1 eV compared with evaluated libraries and previous experimental results.

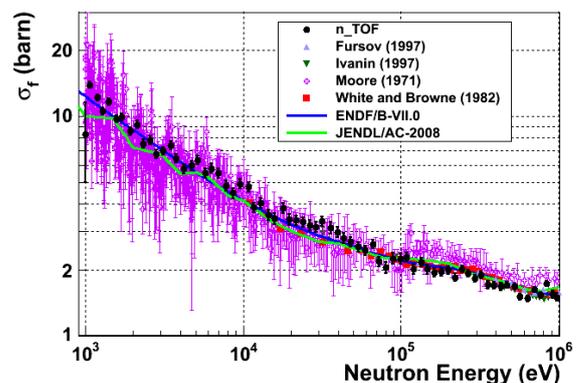


Fig. 4. (Color online)  $^{245}\text{Cm}(n,f)$  n\_TOF data between 1 keV and 1 MeV, compared with evaluated libraries and previous experimental data.

Browne *et al.* [13]. The discrepancy with ENDF/B-VII.0 close to thermal is about 10%. Figure 4 shows the results between 1 keV and 1 MeV: in this energy range n\_TOF data are in average within 2% from ENDF/B-VII.0 and 3% from Moore *et al.*. The systematic uncertainty of the n\_TOF data, including the value induced by the normalization, accounts to about 5% in the full neutron energy range.

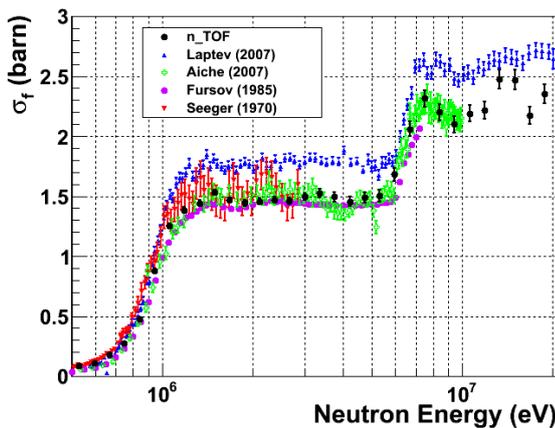


Fig. 5. (Color online)  $^{243}\text{Am}(n,f)$  cross-section measured at n\_TOF around the fission threshold, compared with ENDF/B-VII.0 and previous experimental results.

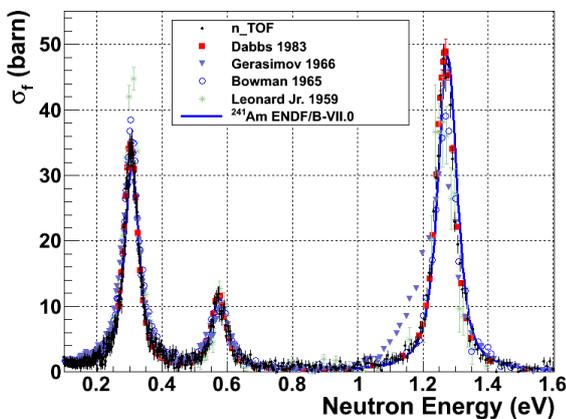


Fig. 6. (Color online)  $^{241}\text{Am}(n,f)$  cross-section between 0.1 and 1.6 eV, compared with previous data and ENDF/B-VII.0.

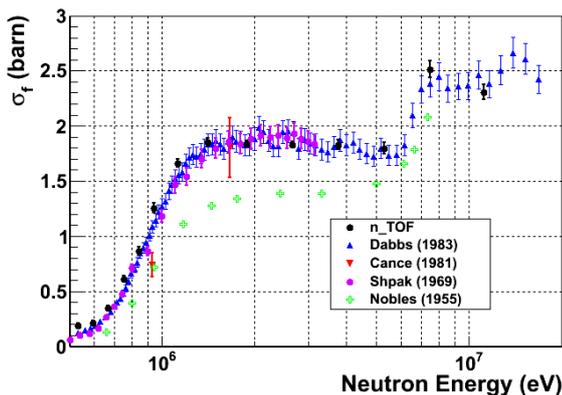


Fig. 7. (Color online)  $^{241}\text{Am}(n,f)$  cross-section around the fission threshold.

An interesting results is obtained for the  $^{243}\text{Am}(n,f)$  reaction, for which fission data are needed to clarify a long standing 15% discrepancy between experimental results. Since, in this case, the background allows the use of a low amplitude threshold, a much more accurate estimate of the efficiency is possible. Therefore, similarly to

the  $^{233}\text{U}$  case, the n\_TOF data are not normalized to previous measurement and a systematic uncertainty around 5% over the full energy range is obtained. Figure 5 shows the n\_TOF data from 500 keV to 20 MeV compared to ENDF/B-VII.0 and to previous results. The experimental points clearly show that the recent data by Laptev *et al.* [14] overestimate the cross-section, while confirming the evaluations of ENDF/B-VII.0. A similar trend was found by Aiche *et al.* [15], in a quasi-absolute measurement. The present dataset therefore now provide strong evidence of the validity of the current evaluated data.

Measurement of the fission cross-section of  $^{241}\text{Am}$  are difficult due to the severe  $\alpha$ -particle background and due to a  $^{239}\text{Pu}$  contamination. With the aim of reducing the uncertainty, the background and  $^{239}\text{Pu}$  subtracted yield were renormalized to the results of Dabbs *et al.* [16] around the 1.27 eV resonance, in a region where the cross-section is known with a reasonable accuracy. The systematic uncertainties are estimated around 10%, mainly due to normalization-related effects. Figure 6 shows the n\_TOF data in the energy region between 0.1 and 1.6 eV around the region where the normalization is performed. Figure 7 shows the data around the fission threshold, in the neutron energy range between 500 keV and 20 MeV. A different analysis procedure was used in this case, similarly to the case of high energy  $^{233}\text{U}(n,f)$  data. It should be noted that, due to the low influence of the  $\alpha$ -particle background in this energy region, it was possible to obtain an absolute value of the cross-section without the need of renormalization to previous data. The n\_TOF results show a reasonable agreement with Dabbs *et al.* data, although more statistics is needed for a more detailed comparison.

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