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Neutron-induced fission cross-section of ^{233}U, ^{241}Am and ^{243}Am in the energy range 0.5 MeV ≤ E_n ≤ 20 MeV

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Abstract

Neutron-induced fission cross-sections of ^{233}U, ^{241}Am and ^{243}Am relative to ^{235}U have been measured in a wide energy range at the neutron time of flight facility n_TOF in Geneva to address the present discrepancies in evaluated and experimental databases for reactions and isotopes relevant for transmutation and new generation fast reactors.

A dedicated fast ionization chamber was used. Each isotope was mounted in a different cell of the modular detector.

The measurements took advantage of the characteristics of the n_TOF installation. Its intrinsically low background, coupled to its high instantaneous neutron flux, results in high accuracy data. Its wide energy neutron spectrum helps to reduce systematic uncertainties due to energy-domain matching problems while the 185 m flight path and a 6 ns pulse width assure an excellent energy resolution.

This paper presents results obtained between 500 keV and 20 MeV neutron energy.

Introduction

Precise neutron-induced fission cross-sections of actinides are required for the design of systems based on the Th/U fuel cycle, for ADS, and Gen-IV nuclear reactors. Requests concerning the accuracy of σ(n,f) data are issued by the OECD/NEA nuclear science committee for some isotopes and reactor types [1].

An extensive measurement campaign for reducing the σ(n,f) uncertainties for major and minor actinide isotopes has been carried out at the n_TOF neutron time of flight facility. In this contribution we report on the ^{233}U, ^{241}Am and ^{243}Am (n,f) cross-sections from 500 keV up to 20 MeV.

Experimental set-up

The n_TOF facility

The n_TOF (Neutron Time Of Flight) facility at CERN is based on a spallation neutron source, consisting of a $80 \times 80 \times 60$ cm^3 thick lead target, which is hit by a pulsed beam of 20 GeV/c protons with 6 ns r.m.s. and a typical repetition rate of 2.4 seconds.

A 185 m long evacuated beam pipe connects the target with the experimental area equipped with several detectors, assuring high energy resolution. The instantaneous neutron flux of 10^5 n/cm^2/pulse at the sample position makes the installation particularly suitable for high accuracy (n,f) cross-section measurements because of the favourable signal to noise ratio related to the natural alpha radioactivity of most actinides. A detailed description of the facility can be found for example in ref. [2] and references therein.

The detector

Neutron-induced fission cross-section measurements have been performed using a Fast Ionization Chamber (FIC). The detector is a modular set of cells. Each cell is composed of three aluminium electrodes 12 cm in diameter which are separated by gaps of 5 mm filled with gas (90% Ar + 10% CF_4) at a pressure of 720 mbar. The central electrode is 100 m in thickness and is plated on both sides with a fissile isotope matching the beam diameter, while the two outer electrodes are 15 m thick. An electric field of 600 V/m is obtained in the gaps by connecting the central electrode to the bias voltage and by keeping the outer ones at ground potential.

Data analysis

Cross-sections are extracted relative to ^{235}U, which is a standard between 0.15 MeV and 200 MeV. At high energy, the output signals of the FIC are strongly affected by the γ-flash, *i.e.* photons and other relativistic particles created in the spallation reaction. A software compensation technique [3] that subtracts the output of two adjacent electrodes was applied to extract the signals of fission fragments. These were further subjected to a pulse shape analysis routine and an amplitude threshold selection to discriminate between fission fragments and α particles.

The TOF information was converted to a neutron energy scale by defining the so-called "time zero" by means of the γ-flash. The "stopping time" was given by the fission fragment signals. The neutron-induced fission cross-sections are extracted according to Eq. (1):

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

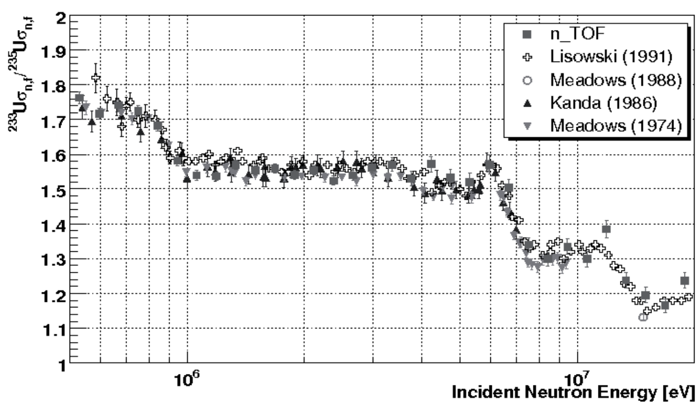
where $\sigma_{235(n,f)}$ is the tabulated ENDF/B-VII.0 cross-section, x stands for the investigated isotope (^{233}U, ^{243}Am or ^{241}Am), N_x denotes the number of fission events detected for isotope x, m_x is the mass (in grams) of isotope x, A_x is the atomic number of isotope x and c_f is a correction factor accounting for dead time effects and detection efficiency. The dead time was treated as non-paralyzable, and the detection efficiency was estimated by simulating the energy loss of fragments in the gas with the FLUKA [4] code. Both corrections are of a few percent only and contribute less than 1% to the total uncertainty.

Considering all effects and corrections introduced in the measurement, the overall systematic uncertainty of the extracted cross-section is 3%. An important contribution is due to the uncertainty of the mass of the various deposits, which is 1.35% for ^{235}U and 1.2% for all other isotopes. The statistical uncertainty is less than 2% for ^{233}U in the whole energy range for a binning of 20 bins/decade and less than 3.5% in the case of ^{243}Am (same binning) for neutron energies higher than 1 MeV. At lower energies, below the fission threshold, the poor counting statistics gives rise to a maximum uncertainty of 7.2%. A variable bin size was used to extract the neutron-induced fission cross-section of ^{241}Am, where the statistical uncertainty is $<2.9\%$ for neutron energies above 1 MeV, and up to 9.5% below the fission threshold.

Results

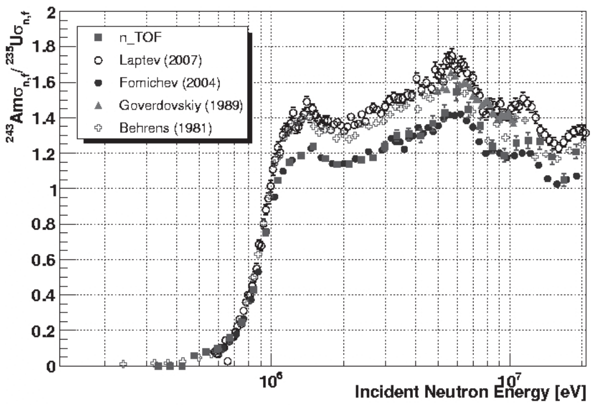
The n_TOF results for ^{233}U agree within 2.1% with the experimental data by Lisowski *et al.* [5], Meadows *et al.*[6] and Kanda *et al.* [7] (see Figure 1). The ENDF/B-VII.0 library is mainly based on these data, though for normalization purposes a "higher ^{235}U $\sigma_{(n,f)}$ was used to produce better agreement with fast critical benchmark experiments" [8]. As a result, at $E_n < 0.7$ MeV we confirm the experimental data reported in Figure 1 and the older ENDF-BVI.8 library, suggesting a revision of the ENDF/BVII.0 evaluation.

Figure 1: Comparison among previous data and n_TOF results for ^{233}U



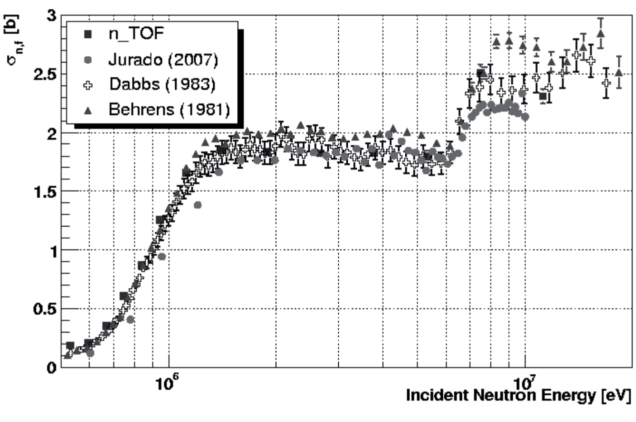
The fission cross-section data for ^{243}Am tend to cluster in two distinct groups in the 1-6 MeV neutron energy range, separated by about 20% from each other (see Figure 2). The Laptev *et al* [9] experimental data confirms the Goverdovskiy *et al.* [10] and the Behrens *et al.* [11], lying in the high energy cluster. The most recent experimental data set at the time of the ENDF/B-VII.0 release was by Laptev *et al.* [9] (information from [8]). Nevertheless such high cross-section values are in disagreement with the averaged cross-sections obtained in the ZEBRA reactor experiment [12], probably the reason why they have been discarded in the ENDF/B-VII.0 evaluation. The n_TOF results confirm Fomichev *at al.* [13] and therefore the low energy cluster.

Figure 2: Comparison among previous data and n_TOF results for ^{243}Am



In the case of the ^{241}Am (n,f) cross-section the most recent experiment reported in EXFOR and ranging up to energies higher than 10 MeV is dated 1983 (Dabbs *et al.* [14]). The n_TOF results agree rather well (within 3%) with this cross-section, confirming this data set rather than the older one by Behrens *et al.* [15]. Jurado *et al* [16] experimental data suggest even lower cross-section values around 10 MeV (see Figure 3).

Figure 3: Comparison among previous and n_TOF results for ^{241}Am



Conclusions

Taking advantage of the high instantaneous neutron flux, the excellent resolution in neutron energy, and the low background of the n_TOF facility, neutron induced fission cross-sections of ^{233}U, ^{241}Am and ^{243}Am have been measured with high accuracy over a wide neutron energy range.

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