NEUTRON-INDUCED FISSION STUDIES AT NCSR "DEMOKRITOS" BY THE NTUA

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Abstract

The neutron beam facility of the 5.5 MV Tandem T11/25 Accelerator Laboratory of the NCSR "Demokritos" has been extensively used over the past 10 years for fission cross section measurements on various actinides (^{237}Np, ^{234}U, ^{236}U, ^{232}Th), at and above the fission threshold. All these isotopes are very important for the design of advanced nuclear systems for a more clean and safe future energy production, as well as for the dissemination of nuclear waste. The neutron beam is produced via the ^{7}Li(p, n), the ^{3}H(p,n), the ^{2}H(d,n) and the ^{3}H(p,n) reactions, depending on the energy range of interest. The neutron flux (of typically 10^5-10^6 n/cm^2/s) is calculated by means of the reference ^{235}U(n,f) and ^{238}U(n,f) cross sections. Special attention is given to the study of the neutron beam (monochromaticity, propagation of neutron beam among the targets etc.), due to the lack of effective threshold for the fission cross section, via detailed Monte Carlo simulations and experimental checks. The detection system consists of a stack of ionization gas cells based on the Micromegas Microbulk technology for the detection of the fission fragments. The final experimental points, which are made publicly available at the scientific community via the EXFOR database, have low uncertainties of the order of 5%. An overview of the experimental campaign, the description of the setup and the analysis as well as the future perspectives will be presented and discussed.

1. INTRODUCTION

Accurate data on neutron-induced reactions are required for the study and development of new generation nuclear systems and alternative fuel cycles, with the scope of making the production of energy through nuclear power economical, proliferation resistant, safer and sustainable. In addition, the study of fission cross-sections on actinides acts as a baseline for the advance and development of the theoretical nuclear models of fission, with the scope to understand the fission process and study the fission characteristics.

In this framework, over the past 10 years, an extensive study of neutron-induced fission cross-sections at and above the fission threshold of various actinides has been carried out at the neutron beam facility of the National Centre for Scientific Research "Demokritos" by the Nuclear Physics Group of the National Technical University of Athens. The measurements were carried out with quasi-monoenergetic neutron beams produced via charged particle reactions on solid and gas targets, while the detection of the fission fragments was achieved with the use of Micromegas detectors. Special attention was given to the estimation of the parasitic neutrons present in the

experimental area, through experimental techniques, as well as Monte Carlo simulations of the neutron beam and experimental setup. The experimental data of the measurements are published and available in the Experimental Nuclear Reaction Data library (EXFOR) [1]. An overview of the above-mentioned measurements and analysis techniques is presented in the paper.

2. EXPERIMENTAL SETUP

The experiments were performed at the 5.5 MV Van de Graaf Tandem Accelerator of the National Centre for Scientific Research "Demokritos" over the past ten years. The production of the neutron beams was achieved via the ^{7}Li(p,n), ^{3}H(p,n), ^{2}H(d,n) and ^{3}H(d,n) reactions, depending on the energy range of interest in each particular measurement, achieving this way cross-section points from the fission threshold up to 18 MeV. The ^{235}U(n.f) and ^{238}U(n,f) cross-sections were used as reference, in order to estimate the neutron fluence incident in the targets. However, it is important to note that the ^{235}U is affected by low energy parasitic neutrons present in the experimental area, thus it was used as reference only in the cases where the contribution of these parasitic neutrons was considered to be negligible.

The targets used in the experiments (^{232}Th, ^{234}U, ^{236}U, ^{237}Np) were characterized by alpha spectroscopy, implementing silicon surface barriers detectors. More specifically, the setup was calibrated by a ^{241}Am alpha source prior to the target measurements [2]. Then, when necessary, additional FLUKA simulations [3] were performed in order to deconvolute the alpha peak of interest from the contaminants present in the target [4], as seen in Fig. 1. In some cases, additional Rutherford Backscattering measurements were performed in order to estimate the homogeneity of the targets [5].

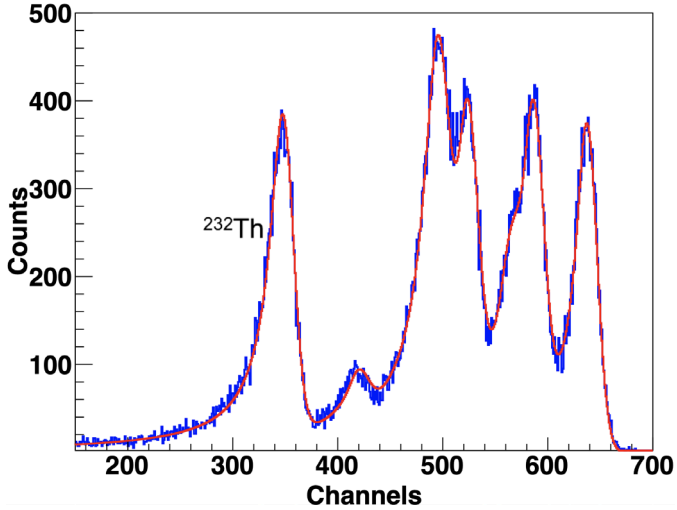

FIG. 1: Experimental alpha spectroscopy spectrum (blue line) along with the FLUKA simulations (red line), where the first peak is the ^{232}Th alpha peak and the rest of the peaks originate from the daughter nuclei.

For the detection of the fission fragments a setup based on the Micromegas gas detectors was used [6, 7]. Each target, acting as the drift electrode, was coupled to a Micromegas detector. From each fission event, one of the created fission fragments enters the detector gas and it is detected, through the energy it deposits in the detector gas. The efficiency of the detector is ~1, while the covered solid angle ~2π. The signal created is collected, after multiplied by avalanches in the mesh region, by a fast preamplifier and it is then fed to conventional electronic modules (amplifier, MCA).

3. DATA ANALYSIS

The neutron-induced fission cross-section at each neutron energy was estimated via the following expression

$$\sigma(E) = \frac{C_{tar}(E)}{C_{ref}(E)} \cdot \frac{\Phi_{ref}(E)}{\Phi_{tar}(E)} \cdot \frac{N_{ref}}{N_{tar}} \cdot \sigma_{ref}(E) \cdot \frac{f_{tar}(E)}{f_{ref}(E)}$$

where C are the counts estimated from the amplitude spectra, Φ is the fluence incident in the target, N is the areal density of the target, σ_{ref} is the cross-section of the reference target and f are various correction factors applied

to the estimated counts that include the dead time correction, correction for the amplitude cut introduced in the analysis to avoid counting alpha counts as fission fragments, correction for the difference in the fluence between the measuring target and the reference target mainly due to the different position with respect to the neutron beam and correction for the parasitic neutrons present in the experimental area, while the subscripts "*tar*" and "*ref*" refer to the measured and reference target respectively.

More specifically, the counts at each neutron energy and each target are estimated from the integration of the spectrum, after applying a suitable amplitude cut to reject the α-particles from the natural radioactivity of the actinide targets present in the low amplitudes of the spectrum. Then, in order to account for the lost fission fragment signals under the α peak, Monte Carlo simulations were performed with the FLUKA code, along with the GEF code [8] to provide the information regarding the energy and mass of the fission fragments of each target, in order to estimate the energy deposition of the fission fragments in the detector gas. The simulated energy deposition spectrum was then calibrated and convoluted with an appropriate function in order to reproduce the experimental one. Thus, the lost fission fragments could be estimated with high accuracy. The experimental and simulated spectra are shown in Fig. 2, where a very good agreement between the two is observed.

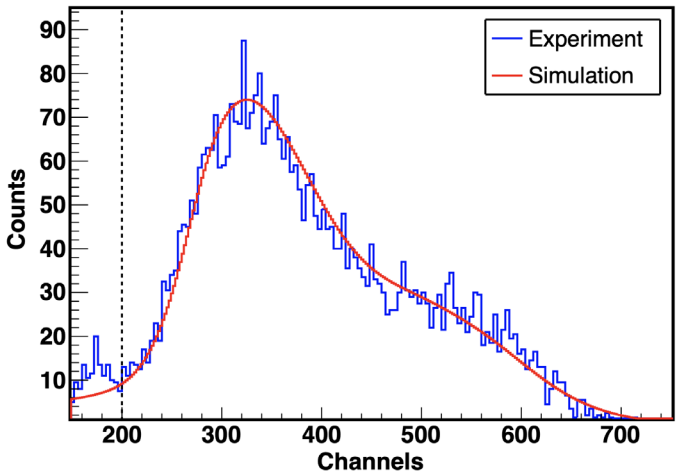

FIG. 2: Experimental fission spectrum (blue line) along with the simulated one (red line) after calibration and convolution with appropriate function. The black dashed line represents the amplitude cut introduced in the analysis.

Additional Monte Carlo simulations were performed with the MCNP code [9], along with the NeuSDesc code [10] for the description of the neutron source, in order to estimate the neutron fluence incident at each target. This is mainly a geometrical correction, originating from the different distances of the targets with respect to the neutron source. Additionally, from these simulations an estimation and correction for low energy parasitic neutrons can be made, by convoluting the flux incident at each target with the reference cross-section of the target [2, 11]. In addition, for the ^{2}H(d,n) reaction the deuteron break-up in the deuterium gas is also taken into account by the simulations. In Fig. 3 the neutron fluence estimated at 10 MeV for the ^{2}H(d,n) neutron producing reaction is presented, where the break-up peak is also present in the spectrum.

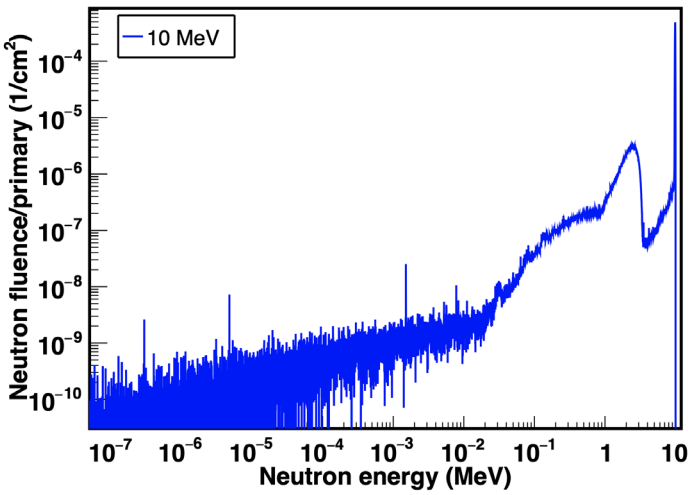

FIG. 3: Simulated neutron fluence for 10 MeV neutrons produced via the ^{2}H(d,n) reaction. The neutron break-up peak is apparent in the spectrum.

In parallel, a thorough study regarding the higher energy parasitic neutrons present in the experimental area has been conducted [3]. These parasitic neutrons originate from interactions of the particle beam with materials in the beam line (^{12}C, ^{16}O, etc.) and the target containers, as well as materials present on the target itself. In order to ensure accurate cross-section results, the similarity in the shape of the cross-section between the reference target and the measuring target, in the regions where these high energy parasitic neutrons are created, is important in order to minimize the effect of these parasitic neutrons. For each reaction measured, an analysis of the effect of the parasitic neutrons is made, depending on the neutron producing reaction and, on the reference, and measuring target. Additional experimental techniques are implemented, as the gasin/gasout method for the $^{2}H(d,n)$ reaction, where measurements are taken for the same neutron energy with and without the deuterium gas, in order to estimate the contribution of the deuterons impinging in the beam line and the materials of the gas cell [2, 5, 6], while the same technique was used for the ^{3}H solid target [3] as well.

4. RESULTS

Cross-section results have been obtained for the neutron-induced fission of

- ^{237}Np in the energy range 4.5-5.3 MeV measured with the $^{2}H(d,n)$ reaction [5].
- ^{234}U in the energy range 400-700 keV, 5.5-10.5 MeV and 14.8-17.8 MeV measured with the $^{7}Li(p,n)$ $^{2}H(d,n)$ and $^{3}H(d,n)$ reactions respectively [2, 12].
- ^{236}U in the energy range 4.5-10 MeV measured with the $^{2}H(d,n)$ [11].
- ^{232}Th in the energy range 2-18 MeV measured with the $^{3}H(p,n)$, $^{2}H(d,n)$ and $^{3}H(d,n)$ reactions [3].

The cross-section data points are available in the EXFOR database [1], while some the latest results concerning the cross-section measurements of ^{232}Th and ^{236}U are presented in Fig. 4 and Fig. 5 respectively.

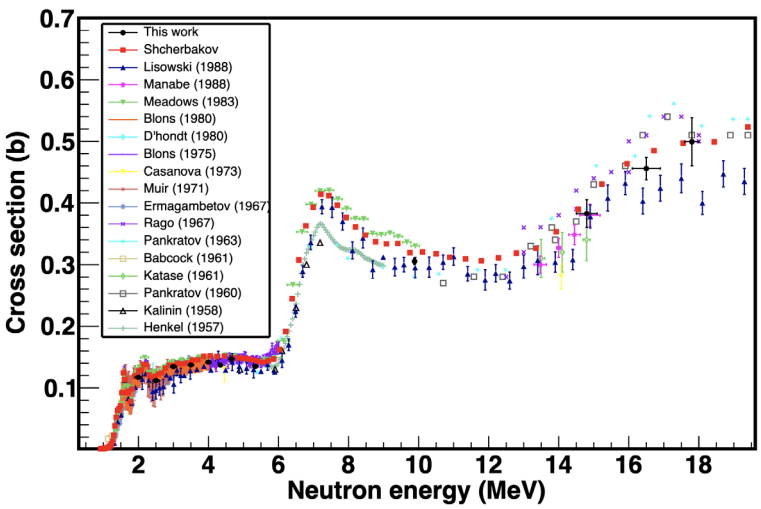

FIG. 4: Cross-section results for the $^{232}Th(n,f)$ reaction (black points) in the energy range 2-18 MeV, along with the available datasets available in EXFOR.

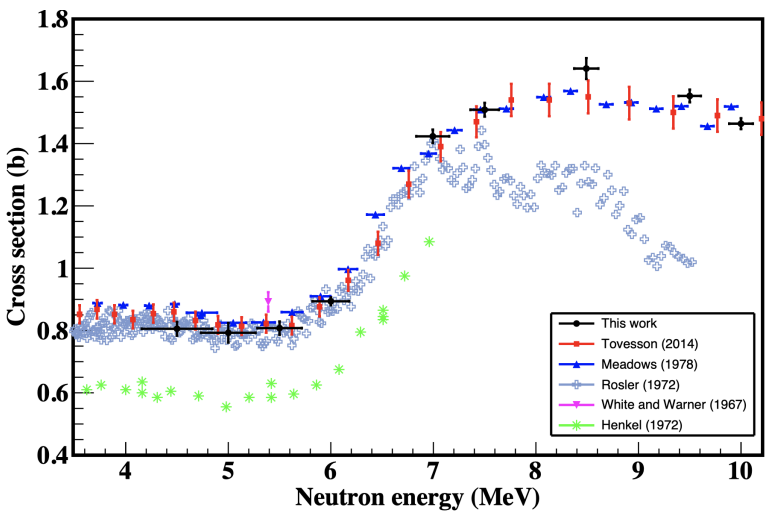

FIG. 5: Cross-section results for the $^{236}U(n,f)$ reaction (black points) in the energy range 4.5-10 MeV, along with the available datasets available in EXFOR.

5. CONCLUSIONS AND FUTURE PERPSPECTIVES

During the last 10 years the Nuclear Physics Group of NTUA has conducted a series of neutron-induced fission cross-section measurements on actinides at the neutron beam facility of the National Centre of Scientific Research "Demokritos". These measurements are compared in the relevant publications with the other existing experimental data and evaluated libraries and are generally found to be in good agreement with certain datasets. The measurements aim to produce data, which can assist in the study and design of advanced nuclear systems and alternative fuel cycles, as well as in the study of the fission process and fission characteristics.

An analysis procedure, which is thorough and detailed, has been developed in order to extract accurate cross-section results, with special attention given to the study of parasitic neutrons based on experimental and simulated techniques. As a next step, Monte Carlo simulations are being performed with the GEANT4 code [13], in order to have an estimation of the interaction of the charged particle beam, with the beamline and target materials, along with the simulation of the neutron beam.

Finally, measurements on the highly radioactive ^{233}U are scheduled to be performed in the neutron beam facility of "Demokritos", which will yield interesting new data to complement the existing results.

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