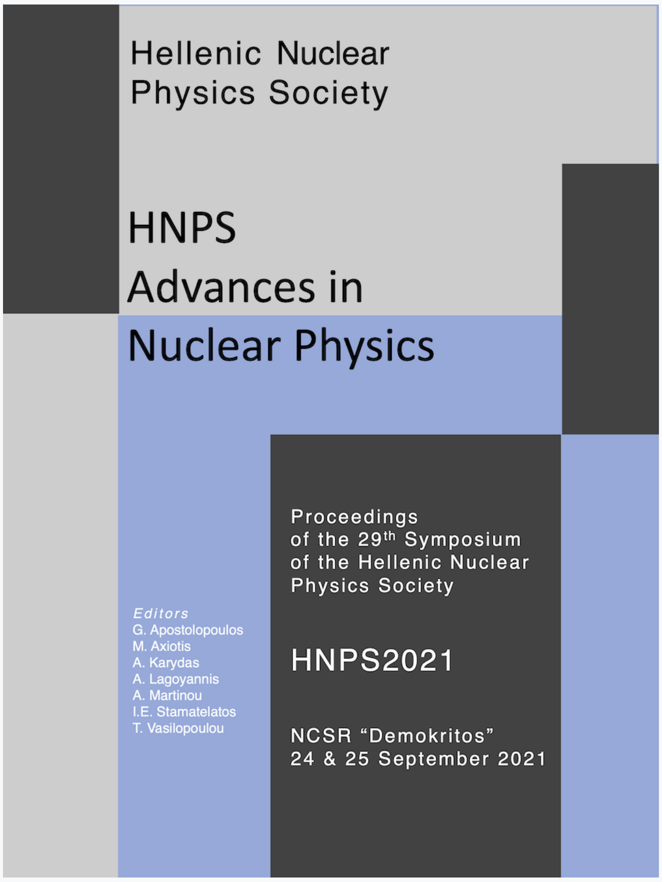
HNPS Advances in Nuclear Physics

Vol 28 (2021)

HNPS2021



Study of the 3H(p,n)3He neutron producing reaction at N.C.S.R. "Demokritos" – Application on the 232Th(n,f) reaction

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doi: [10.12681/hnps.3708](https://doi.org/10.12681/hnps.3708)

Study of the ^{3}H(p,n)^{3}He neutron producing reaction at N.C.S.R. "Demokritos" – Application on the ^{232}Th(n,f) reaction

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Abstract In the present work, the neutron beams produced via the ^3H(p,n) ^3He reaction, were studied at N.C.S.R. "Demokritos". For detecting and monitoring the neutrons, the following reference reactions ^{238}U(n,f), ^{235}U(n,f) and ^{237}Np(n,f) were used. Furthermore, a systematic study of the parasitic neutrons, produced via reactions on the target constituents, was performed. At the same time, the cross sections of the ^{232}Th(n,f) reaction were deduced, in the energy range from 2 to 5.5 MeV. Seven actinide targets were used, coupled with seven Micromegas detectors, one for each target, for the detection of the fission fragments. The target-detector assembly was placed in an aluminum chamber filled with Ar:CO_2 (90:10) in atmospheric pressure and room temperature. Monte Carlo simulations with the MCNP6 code, coupled with the NeuSDesc and SRIM-2013 codes, were used for the estimation of the neutron beam incident at each target. Additional Monte Carlo simulations were carried out using the codes FLUKA and GEF, in order to determine the exact masses of the ^{232}Th targets and the energy deposition of the fission fragments in the detector gas.

Keywords neutron beams, parasitic neutrons, fission, cross section

INTRODUCTION

Studies of neutron induced reactions are of considerable interest, not only for their importance to fundamental research in Nuclear Physics, but also for practical applications such as reactor technology. The study of these reactions is one of the main research interests of the Nuclear Physics Group at the National Technical University of Athens. One of the laboratories used for the experimental measurements of these reactions, is the Institute of Nuclear and Particle Physics of N.C.S.R. "Demokritos", at the 5.5 MV Tandem Van de Graaff accelerator. For the production of the neutron beams, several charged particle reactions on solid and gas targets are being used, covering a wide neutron energy range [1]. In this framework, the ^{3}H(p,n)^{3}He reaction was used for the first time at N.C.S.R. "Demokritos" in order to produce quasi-monoenergetic neutron beams. Part of the aim of the present work was to study the characteristics of the produced neutron beams in various energies.

In parallel, the neutron induced fission cross section of ^{232}Th was measured, using the neutron beams that were produced via the ^{3}H(p,n)^{3}He reaction. The accurate cross-section data of neutron induced fission on actinides are of considerable importance both for the design of advanced nuclear systems, such as Generation IV reactors and Accelerator-Driven Systems (ADS), as well as for the study of the fission process itself [2-3]. In addition, since ^{232}Th is 3 to 4 times more abundant than uranium, the thorium fuel cycle is considered an alternative to the conventional uranium cycle [4-6]. This part of the work aims to resolve the discrepancies in the energy range from 2 to 5.5 MeV, where

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differences up to 11% between the different evaluation data sets [13, 18-19] and 30% between the experimental ones [16-17] have been observed.

EXPERIMENTAL DETAILS

The experiment was carried out at the 5.5 MV Tandem Van de Graaff accelerator of N.C.S.R. "Demokritos". For the production of the neutron beams, a solid Ti-tritiated target of 373 GBq activity was used. The target consisted of a 2.1 mg/cm^2 Ti-T layer with a 25.4 mm diameter, placed on a 1 mm thick Cu backing with a 28.5 diameter, and in front of the Ti-T target there was a 10 μm Mo foil.

Seven actinide targets were used in total, two ^{232}Th, two ^{238}U and one ^{235}U, ^{234}U and ^{237}Np. The samples were produced at the IPPE (Obnisk) and JINR (Dubna) via the painting technique. The actinide material was deposited on a 100 μm Al backing. During the irradiations, aluminum masks of 0.6 mm thickness and 5 cm diameter were placed in front of each target in order to define the angular acceptance of the neutron beam and minimize the uncertainty of the neutron energy.

One of the most important characteristics of the actinide targets is the number of nuclei. To accurately determine this parameter, α-spectroscopy measurements were carried out, using a Silicon Surface Barrier (SSB) detector. Each target along with the aluminum mask was placed in a vacuum chamber, at a distance of 0.1 cm from the silicon detector. From the obtained data, the number of nuclei in each actinide target was estimated using the following equation:

$$N = C_{\Omega} \cdot \frac{4\pi}{\Omega} \cdot \frac{t_{1/2}}{ln2}$$

where C_Ω are the α-particles measured by the silicon detector per second, Ω is the solid angle between the actinide target and the silicon detector and $t_{1/2}$ is the half-life of the actinide target in seconds [7]. Monte Carlo simulations were performed with the FLUKA code [26-27] in order to estimate the tail in which the ^{232}Th α peak lies, which originates from its daughter nuclides that are also α emitters.
For the detection of the fission fragments seven different Micromegas detectors [8] were used, one for each target. The whole target-Micromegas setup was placed in an aluminum chamber filled with a gas mixture of Ar:CO_2 (90:10) kept at atmospheric pressure and room temperature. In Fig. 1, the target-detector assembly along with the aluminum chamber are presented.

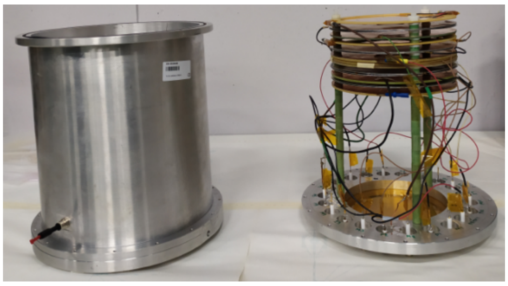

Fig. 1. *The aluminum chamber and the target-detector assembly.*

MONTE CARLO SIMULATIONS

In order to determine the neutron fluence incident on each target, Monte Carlo simulations with the MCNP6 code [9] were performed. A detailed description of the geometry of the experiment was used, while the NeuSDesc [25] code was implemented for the description of the neutron source imported in MCNP6, giving all the necessary input information (reaction, ion energy, entrance foil thickness). The NeuSDesc code creates a detailed description of the neutron source taking into account the energy loss of the proton beam in the Mo foils via SRIM-2013 [10] calculations and all the kinematic parameters of the ^{3}H(p,n)^{3}He reaction. Except from the main neutron beam, parasitic neutrons produced via scattering on the experimental setup are also included in the simulations. The neutron fluence estimated from the Monte Carlo simulations for the first target (^{238}U) for the irradiation corresponding to neutron energy 3 MeV is presented in Fig 2.

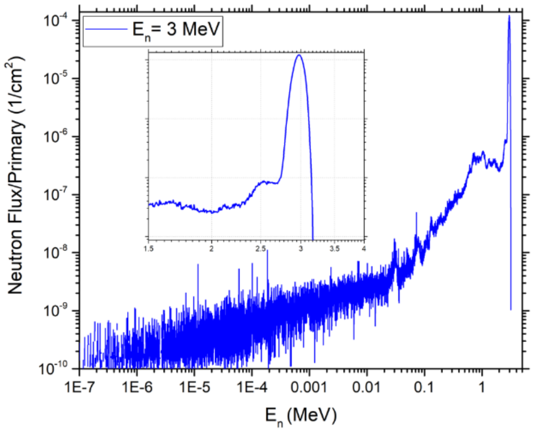

Fig. 2. *Simulated neutron fluence from MCNP6 on the ^{238}U sample for the irradiation at 3 MeV. The neutron fluence from the main peak is shown in the inset.*

THE ^{3}H(p,n)^{3}He REACTION

In total eight irradiations were performed, using eight proton beams from 3.4 to 6.5 MeV, a region where the ^{3}H(p,n) reaction cross section has its highest values. These proton beams correspond to eight different quasi-monoenergetic neutron beams in the energy range between 2 and 5.5 MeV.

One of the basic characteristics of this reaction, that was crucial to study before the beginning of the irradiations was the angular distribution for both the cross section and the energy of the produced neutrons. For the determination of the cross section angular distribution the methodology described by H. Liskien et al. [11] was adapted, while for the energy of the produced neutrons, kinematic calculations were performed. After reviewing the results, the fission chamber was placed in a 2.5 cm distance from the neutron source, where the angular acceptance of the first actinide target was ±10º, range where the neutron beam can be considered isotropic and quasi-monoenergetic.

As mentioned above, the tritium target consisted of multiple materials (Ti, Cu, Mo). These materials can be considered as potential "targets" for the production of parasitic neutrons via (p,n) reactions [12]. For monitoring the neutron beam and checking the presence of parasitic neutrons during the irradiations, the three following reference reactions ^{238}U(n,f), ^{235}U(n,f) and ^{237}Np(n,f) were used. In order to find out if parasitic neutrons were present during the irradiations, two different sets of cross section calculations were carried out, one for the ^{238}U(n,f) reaction using as reference the ^{235}U(n,f) reaction and the second one for the ^{237}Np(n,f) reaction using as reference the ^{238}U(n,f) reaction. The target-reference combinations were chosen in a way where the detection of both thermal and fast parasitic neutrons was possible due to the different cross-section behavior of each reaction with respect to neutron energy.

After completing the calculations, the experimental results were compared with the proposed values of the latest ENDF/B-VIII.0 evaluation [13]. It was observed that for the first two irradiation cycles (E_n = 2.0 and 2.5 MeV) the experimental results were in agreement with the evaluated ones, within their statistical uncertainties. After the third irradiation, discrepancies between the experimental values of both reactions and the ones from ENDF/B-VIII.0 started to appear. In the case of the ^{238}U(n,f) reaction these discrepancies ranged from 20% at the third irradiation, up to 124% at the highest neutron energy, while in the case of the ^{237}Np(n,f) reaction these discrepancies ranged from 6% to 86%, respectively.

These differences confirmed the fact that after the second irradiation parasitic neutrons were generated via (p,n) reactions at the target's materials. To check which (p,n) reaction was activated, on which one of the stable isotopes of Ti, Cu, Mo and during which irradiation, five parameters were taken into account: the energy of the proton beam before impinging on each material, the Q_{value}, the threshold energy, the Coulomb barrier and the cross section of each reaction. After analyzing these parameters, it was realized that from the third irradiation, (p,n) reactions on Ti isotopes were activated and as the proton beam increased in energy, other (p,n) reactions on various isotopes of Ti, Cu and Mo were additionally activated, generating parasitic neutrons in different energy ranges.

In order to estimate the effect of these parasitic neutrons, calculations were performed to approximate the parasitic peaks from the various reactions, taking into account the cross section of each reaction at the different proton energies as they were given by TENDL-2019 [14], an approximate mass for the nuclei of the materials and the fluence of the proton beam, estimated via the current integrator. Unfortunately, a quantitative result was not possible, but it was observed that the addition of the parasitic neutrons on the total neutron fluence determined by MCNP6, was able to decrease the discrepancies between the experimental and the evaluated data.

The analysis described above, concluded that the $^{3}H(p,n)^{3}He$ reaction, considering the N.C.S.R. "Demokritos" experimental setup, can be used for the production of monoenergetic neutron beams with energies up to 2.5 MeV. At higher energies, various (p,n) reactions on the target materials are generating parasitic neutrons, so this reaction must be used with caution for cross-section measurements, taking into account the specific needs and characteristics of each experiment.

^{232}Th FISSION RESULTS AND DISCUSSION

The cross section of the ^{232}Th(n,f) reaction is estimated by the expression:

$$\sigma_{^{232}Th} = \frac{(Y \cdot f_{amp} \cdot f_{par} \cdot f_{DT})_{^{232}Th}}{(Y \cdot f_{amp} \cdot f_{par} \cdot f_{DT})_{Ref}} \cdot \frac{\Phi_{Ref}}{\Phi_{^{232}Th}} \cdot \frac{N_{Ref}}{N_{^{232}Th}} \cdot \sigma_{Ref}$$

where

- Y is the fission yield recorded by the Micromegas detectors and corrected for: the amplitude cut used in the analysis to reject the alpha counts from the natural radioactivity of the samples (f_{amp}), the dead time (f_{DT}) and the contribution from low energy parasitic neutrons in the fission yield, occurring from scattering of the main neutron beam in the experimental setup (f_{par})
- N is the number of nuclei in the actinide samples
- Φ is the neutron fluence on each sample
- σ_{Ref} is the cross section of the reference target obtained from evaluation libraries

The results for the neutron induced fission cross section values for ^{232}Th, using as reference the ^{238}U(n,f) reaction have already been published by V. Michalopoulou et al. [15] and are shown in Fig. 3, along with other experimental data available in EXFOR [16-17] and existing evaluation libraries [13, 18-19]. Due to the similar fission cross section of ^{232}Th and ^{238}U, the contribution from the parasitic neutrons mentioned above in the final cross section results was negligible.

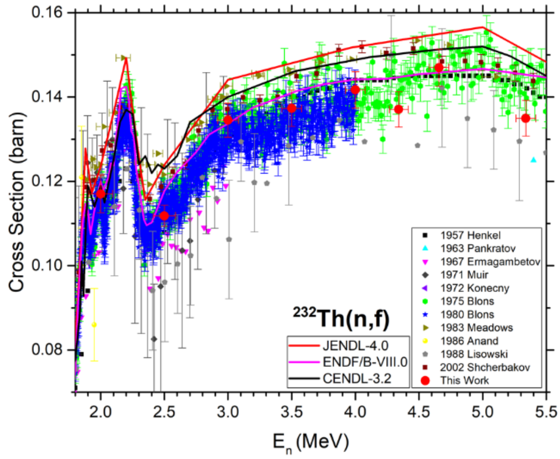

Fig. 3. *The results for the ^{232}Th(n,f) cross section measurement.*

The results are in good agreement with the data of Blons et al. [20-21] and Henkel et al. [22]. Also, there is an agreement within statistical uncertainties with the experimental data of Lisowski et al. [23] and Shcherbakov et al. [24]. Concerning the values from the different evaluation libraries, the present results are in overall good agreement with the latest ENDF/B-VIII.0 evaluation [13].

Acknowledgments

The authors would like to acknowledge the assistance of the Accelerator Staff at N.C.S.R. “Demokritos”. This research is implemented through IKY scholarships program and co-financed by the European Union (European Social Fund –ESF) and Greek national funds through the action entitled “Reinforcement of Postdoctoral Researchers -2nd call (MIS 5033021)”, in the framework of the Operational Programme “Human Resources Development Program, Education and Lifelong Learning” of the National Strategic Reference Framework.

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