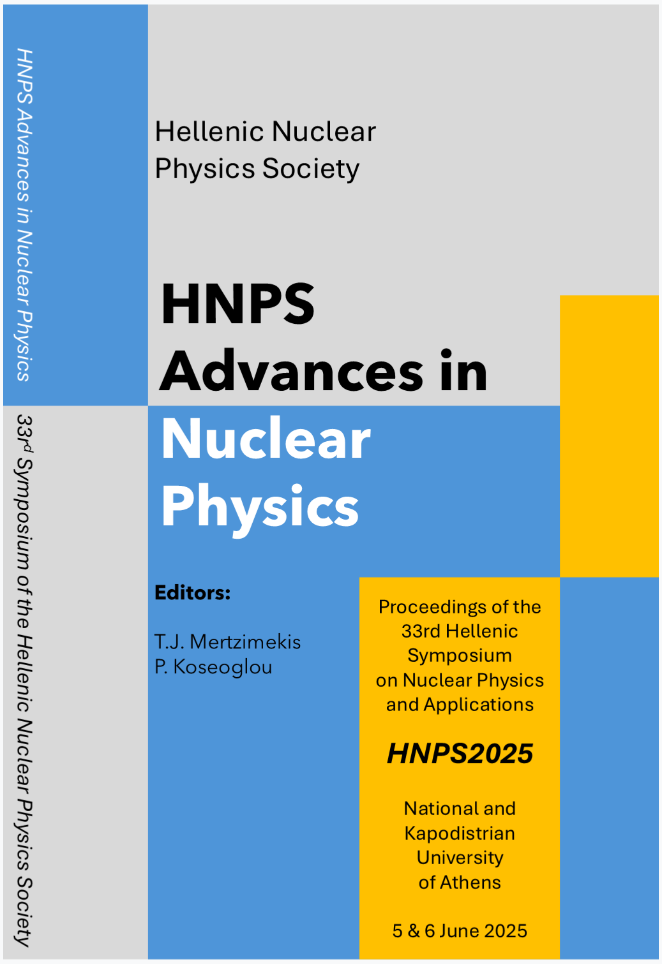
HNPS Advances in Nuclear Physics

Vol 32 (2026)

HNPS2025



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doi: [10.12681/hnpsanp.8961](https://doi.org/10.12681/hnpsanp.8961)

HNPS Advances in Nuclear Physics Vol. **32**, 205–211 (2025)
doi: 10.12681/hnpsanp.8961

Hellenic Nuclear Physics Society
Advances in Nuclear Physics
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ARTICLE

Cross Section Measurements of Neutron Induced Reactions on Mo Isotopes

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(Received: 22 Nov 2025; Accepted: 02 Feb 2026; Published: 03 Feb 2026)

Abstract

The cross section of the ^{100}Mo(n,2n)^{99}Mo reaction has been measured with the activation technique and ^{27}Al(n,α)^{24}Na as a reference reaction, at 16.0 and 14.6 MeV neutron energies. The measurements were performed at N.C.S.R. "Demokritos" during the same irradiation, by placing the target assemblies both at 0° and 80° with respect to the primary deuteron beam that was used for the neutron production via the ^{3}H(d,n)^{4}He reaction. In the present work, the neutron production at 14.6 MeV was thoroughly investigated, both experimentally and via Monte Carlo simulations with the MCNP6 code, as it was used for the first time at N.C.S.R. "Demokritos" for cross section measurements. More specifically, simulations were carried out: the first one to study energy variations with laboratory angle, and the second to replicate this specific experiment using a 2.15 MeV deuteron beam and targets positioned at 0° and 80°, for corresponding to neutron energies of 16.0 and 14.6 MeV, respectively. Additionally both ^{100}Mo(n,2n)^{99}Mo and ^{27}Al(n,α)^{24}Na reactions were employed to validate the simulation results. Good agreement between simulation and experiment demonstrates that the approach reliably reproduces both the angular and energy characteristics of the neutron beam, providing a foundation for further studies at additional neutron energies and angles.

Keywords: neutron activation; MCNP simulations ; angular distribution ; cross section measurements ; Mo

1. Introduction

Neutrons play an important role in scientific research and applications including energy production, medical therapies and study of materials. Depending on the required neutron beam features in the applications, different neutron sources can be used, such as nuclear fission reactors, radioactive isotopes emitting neutrons and accelerator-based neutron sources. In the present work, the neutron beam was produced via the ^{3}H(d,n)^{4}He reaction, which is commonly used in accelerators, producing high-yield, nearly monoenergetic fast neutrons, whose energy depends on the emission angle.

Investigation of the neutron beam characteristics at various energies and directions was carried out by performing Monte Carlo simulations with the MCNP6 code [1]. Two different simulations were performed, the one aimed to investigate the neutron energy variations with laboratory angles and the second was complementary to a specific experimental measurement, carried out in the Institute of Nuclear and Particle Physics of N.C.S.R. "Demokritos", at the 5.5 MV Tandem Van de Graaff accelerator utilizing a E_d = 2.15 MeV deuteron beam. In this experiment targets of Mo and Al were placed in two directions, at 0° and 80° with respect to the beamline, in order to be irradiated with 16.0 and 14.6 MeV neutrons, respectively. Neutron activation analysis (NAA) technique was applied to experimentally determine the cross section of the ^{100}Mo(n,2n)^{99}Mo reaction. The accuracy of the simulated neutron energy angular distribution was validated through comparisons with experimental results obtained from the ^{27}Al(n,α)^{24}Na monitor reaction.

2. The ^{3}H$(d, n)^4$He reaction

The ^{3}H(d,n)^{4}He is an exothermic reaction that its large positive Q-value (Q = 17.59 MeV) and the low atomic numbers involved allow for high yield production (see Fig. 1) of fast, nearly monoenergetic 14-15 MeV neutrons. It can be performed either with neutron generators, by using relatively low deuteron energies, or at accelerator facilities, by using higher deuteron energies. Depending on the emission angle, this reaction produces neutrons with energies ranging from approximately 11.5 to 20.5 MeV [2]. Reaching the extreme ends of this energy range is challenging: the maximum energy is affected by secondary neutrons due to breakup of the projectile via reactions ^{3}H(d,np)^{3}H and ^{3}H(d,2n)^{3}He [3], while achieving the minimum energy is often experimentally difficult. Consequently, examining the angular dependence of the reaction is crucial for comprehending the variation of neutron energy with angle and facilitating irradiation at lower neutron energies.

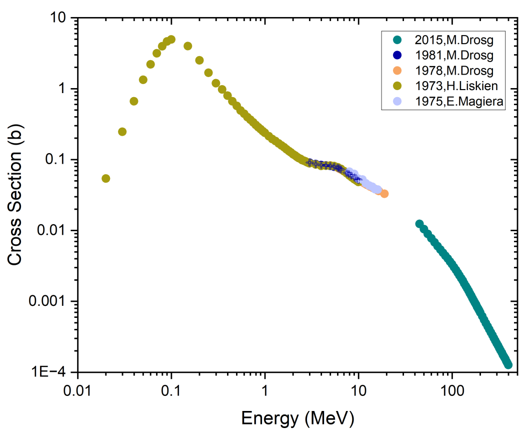


Figure 1. Experimental cross section for the ^{3}H(d,n)^{4}He reaction [2–7]

3. Monte Carlo Simulations

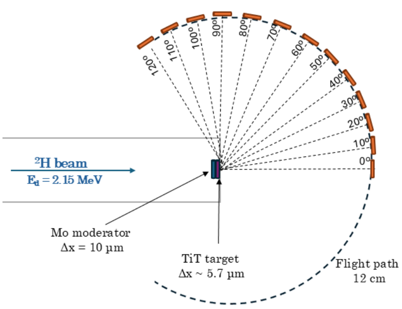
To investigate the angular dependence of the neutron energy in detail, Monte Carlo simulations with the MCNP6.1 code were performed, including the complete experimental beamline geometry of the Institute of Nuclear and Particle Physics of N.C.S.R. "Demokritos", such as TiT target and the

10 μm Mo foils right before the target. The NEBOAS [8] code was implemented for the description of the neutron source imported in MCNP6, giving all the necessary input information (reaction, ion energy, degrader foil thickness). The NEBOAS code generates a detailed description of the neutron source, considering the energy loss of the incident beam in the Mo foils via SRIM-2013 [9] and all relevant kinematic parameters of the ^{3}H(d,n)^{4}He reaction.

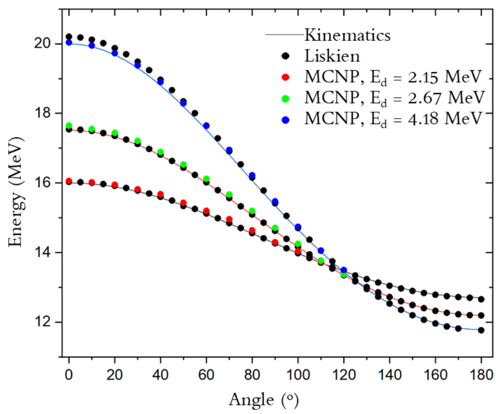
As previously mentioned, two different simulations were carried out. In the first simulation, Al foils with the same geometrical characteristics as those used in the experiment (see next section), were modeled to ensure identical angular acceptance and consistent comparison with the experimental conditions. The foils were arranged in a circular pattern around the TiT target, with a radius of 12 cm and placed at 10° intervals, as shown in Fig. 2a. The results of this simulation provided the angular distribution of the emitted neutron energy, calculating the flux-averaged neutron energy using the following equation:

$$\bar{E} = \frac{\sum \Phi_i E_i}{\sum \Phi_i} \tag{1}$$

with Φ_i representing the simulated neutron flux derived from MCNP6 and E_i the corresponding neutron energy. The calculated energy according to MCNP and NEBOAS coupling simulations simulations showed good agreement with both the experimental data of Liskien and the two-body kinematic calculations for different deuteron, and consequently, neutron energies, as illustrated in Fig. 2b. This procedure has been repeated and examined for various deuteron/neutron energies to verify the agreement in other energies, also, and validate the method. Fig. 3 shows the differential neutron yield as a function of angle and neutron energy in the laboratory system for the case studied in Fig. 2 and for deuteron energy 2.15 MeV. The neutron energy at 0° is 16.0 MeV and the distribution at other angles reflects the expected angular dependence.



(a) Scheme of the simulated setup. The target assembly and the foils are shown.



(b) Comparison between the simulated effective energy, Liskien data and two body kinematics calculation.

Figure 2. Simulation results

Following the verification of the simulation approach through the angular neutron energy study, a second MCNP simulation was performed corresponding to an exact replication of the experimental configuration of the neutron activation experiment, described in the next session. This simulation was not only intended to assess the applicability of the angular study under realistic experimental conditions but also constituted an integral part of the experimental data analysis. The full geometry of the experimental setup was implemented in MCNP, including all relevant materials along with the target assembly used for neutron induced cross section measurements. In this configuration, the neutron flux at the aluminum reference foils was simulated and subsequently compared with the

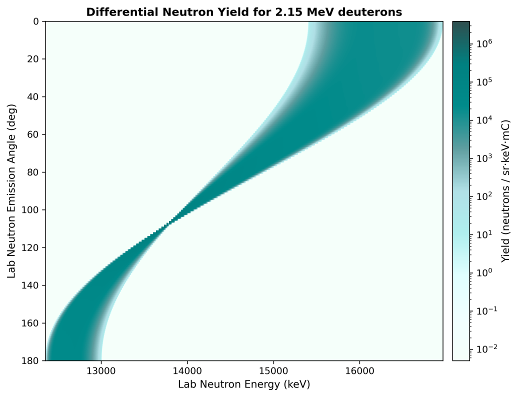


Figure 3. Differential Neutron yield as a function of laboratory angle and neutron energy in the laboratory system, with E_d=2.15 MeV, calculated by the NEBOAS code

experimental measurements, as presented in the following section. This approach allowed for an accurate description of the neutron field at the foil positions while ensuring the consistency of the simulation methodology when applied to the exact experimental setup.

4. Neutron Activation Analysis Experiment

Experimental measurement of the ^{100}Mo(n,2n)^{99}Mo cross section was carried out, that utilized neutron beam generated via the ^{3}He(d,n)^{4}He reaction at the 5.5 MV Tandem Van de Graaff accelerator facility of NCSR "Demokritos". For a deuteron energy of 2.15 MeV, the neutron energies were 16.0 MeV at 0° and 14.6 MeV at 80°. Specifically, two assemblies of natural Mo foils placed between two Al reference foils, were positioned at the target holders placed at 0° and 80°, at distances of 2 cm and 6 cm from the TiT target, respectively, as shown in Fig. 4. In the case of the target assembly at 80°°, two more Al foils and a Tl foil were included as well for a separate cross section measurement of the ^{203}Tl(n,2n)^{2020}Tl. This experimental setup was also designed specifically to assess the consistency and accuracy of the simulation derived neutron energy and flux distributions.

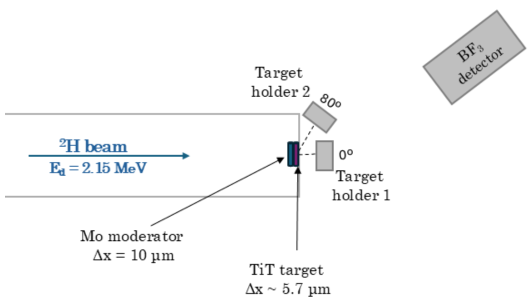


Figure 4. Scheme of the experimental setup

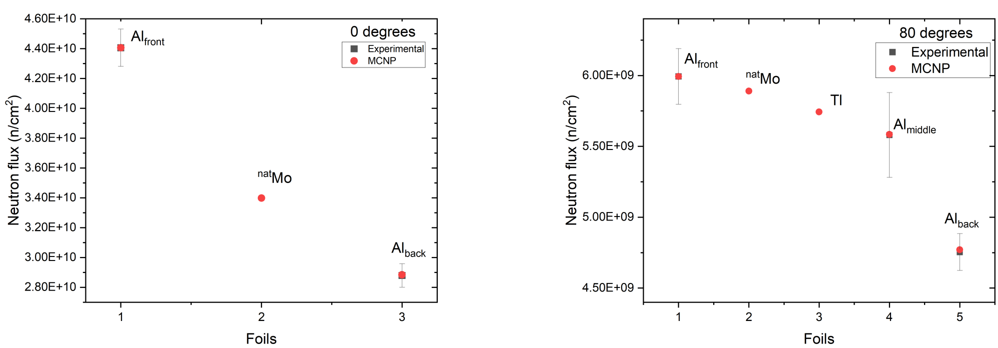
The flux variation of the neutron beam was monitored by a BF_3 detector placed at a distance of 2 m from the neutron producing target. Following the irradiation, the activities of both the Mo targets and Al reference foils were measured offline with HPGe detectors. The absolute neutron flux was determined through the reference reaction ^{27}Al(n,α)^{24}Na, allowing direct comparison with the MCNP predictions and thereby validating the simulation approach. For the cross-section calculation based on the activation technique, the following general expression was used:

$$\sigma_{\text{tar}} = \sigma_{\text{ref}} \cdot \frac{N_{\gamma,\text{tar}}}{N_{\gamma,\text{ref}}} \cdot \frac{(\varepsilon F I_\gamma D f_c N_t)_{\text{ref}}}{(\varepsilon F I_\gamma D f_c N_t)_{\text{tar}}} \cdot \frac{\Phi_{\text{ref}}}{\Phi_{\text{tar}}} \tag{2}$$

The subscripts "ref" and "tar" refer to the reference and measured target, respectively, and the factors in Eq. (2) are listed below [10].

- N_γ: is the integral of the γ-ray peak of interest
- N_t: is the total number of nuclei in the target or the reference foil
- σ: is the cross section
- ϵ: is the absolute efficiency of the HPGe detector at the γ-ray energy of interest
- F: is a correction factor for the self-absorption of a γ-ray within the sample itself
- I_γ: is the γ-ray intensity
- D: is a correction factor regarding the radioactive decay of the nuclei for the time intervals between the end of the irradiation and the start of the measurement
- f_c: represents the correction factor for the nuclei that are decaying during the irradiation time, accounting for potential neutron beam instabilities as well. The integrated counts f(t) are recorded at short intervals dt using a BF_3 detector.
- $\frac{\Phi_{\text{ref}}}{\Phi_{\text{tar}}}$: is the ratio of neutron flux between the measured target and reference foil. It is obtained from MCNP6 simulations.

The full geometry of the experiment was described in MCNP, and the neutron flux at the Al foils was first simulated and then compared with the experimental results. The good agreement between simulation and experiment confirmed that the simulation that replicated the experiment accurately represents the neutron field at the foil positions, as shown in Fig. 5.



(a) Foils at 0°

(b) Foils at 80°

Figure 5. Experimental neutron fluences in the reference foils along with the simulated ones, obtained by means of the MCNP6 code. For both different laboratory angles, the first Al foil was used for normalization.

The neutron energies at the experimental foil positions, obtained from the second simulation, were consistent with the previously modeled beamline, with mean energies of 16.0 and 14.6 MeV and corresponding standard errors 0.3 and 0.4 MeV, respectively. Finally, by using this validated simulation, the cross section of the ^{100}Mo(n,2n)^{99}Mo reaction was calculated at both angles and corresponding neutron energies. The results are in good agreement with the available experimental data [11] and the latest evaluations of ENDF/B-VIII.1, JEFF-3.3, JENDL-5 and TENDL-2023 [12], as shown in Fig. 6. Especially at 14.6 MeV, where many data points exist in literature, with discrepancies among them reaching up to 50%, the cross section value of the present work lies in the region of the mean value and provides crucial information for validating nuclear models and evaluation libraries. This agreement also provides an experimental validation of the simulations and confirms that the MCNP model accurately estimates both the angular and energy characteristics of the emitted neutrons. Furthermore, the exact same method was used for the measurement of ^{203}Tl(n,2n)^{202}Tl reaction under the same irradiation conditions, demonstrating excellent agreement with the current experimental data, thus further validating this approach.

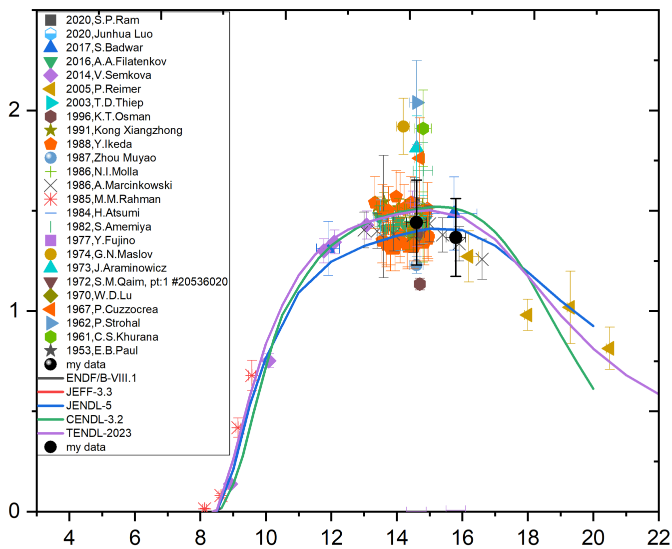


Figure 6. Experimental cross section for the ^{100}Mo(n,2n)^{99}Mo reaction at 14.6 and 16.0 MeV.

5. Conclusion

The cross section of the ^{100}Mo(n,2n)^{99}Mo reaction has been measured with the activation technique at 16.0 and 14.6 MeV. The results are in good agreement with the data available in literature and the latest evaluation libraries. The neutron production at 14.6 MeV has been achieved by placing the target assembly at 80° with respect to the main neutron beam, which was produced via the ^{3}H(d,n)^{4}He reaction. This set-up was used for the first time at N.C.S.R. “Demokritos” for cross section measurements and was thus thoroughly investigated with Monte Carlo simulations using the MCNP6 code to examine the angular and energy dependence of neutrons produced by the ^{3}H(d,n)^{4}He reaction. The simulations demonstrated good agreement with the expected neutron energies when compared to available experimental data for ^{100}Mo(n,2n)^{99}Mo, ^{203}Tl(n,2n)^{202}Tl [13], and ^{27}Al(n,α)^{24}Na reactions. This approach effectively validated the dependability of the simulation methodology, and more experimental studies at different angles, energies, and reactions are anticipated to expand on these findings.

Acknowledgements

This work was co-funded by the European Union under grant agreement 101164596 - APRENDE Project. Views and opinions expressed are however, those of the author(s) only and do not necessarily reflect those of the European Union or the European Atomic Energy Community ('ECEuratom').

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