



The neutron facility at NCSR ‘Demokritos’ and neutron activation research activities of NTUA

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Abstract

At the 5.5 MV Tandem T11/25 Accelerator Laboratory of NCSR “Demokritos” quasi-monoenergetic neutron beams can be produced in the energy ranges $\sim 15\text{--}21$ MeV by means of the ${}^3\text{H}(\text{d},\text{n}){}^4\text{He}$ reaction, $\sim 4\text{--}11$ MeV via the ${}^2\text{H}(\text{d},\text{n}){}^3\text{He}$ reaction, $\sim 2.0\text{--}5.3$ MeV using the ${}^3\text{H}(\text{p},\text{n}){}^3\text{He}$ reaction and $\sim 120\text{--}650$ keV via the ${}^7\text{Li}(\text{p},\text{n}){}^7\text{Be}$ reaction. The maximum flux has been determined to be of the order of $10^5\text{--}10^6$ n/cm²s, implementing reference reactions, while the flux variation of the neutron beam is monitored using a BF₃ detector and a BC501A scintillator. The neutron beams have been characterized using the multiple foil activation technique as well as extensive simulations and have been used for cross section measurements of neutron induced reactions implementing again the activation technique.

1 Introduction

Studies of neutron induced reactions are of considerable interest, not only for their importance to fundamental research in Nuclear Physics and Astrophysics, but also for practical applications in medicine, nuclear technology, dosimetry and industry. The main technological applications are related to the design of innovative Generation-IV reactors and Accelerator Driven Systems (ADS) for the future production of clean and safe nuclear energy as well as for the transmutation of nuclear waste. All these tasks require improved nuclear data and cross sections of high precision for neutron induced reactions on various isotopes. It is thus of major importance that the performance of the neutron sources is well understood and that the experimental conditions are well characterized.

In view of the above remarks and in absence of time-of-flight capabilities, the neutron facility at the Athens 5.5 MV tandem accelerator of NCSR “Demokritos” [1] has been characterised by means of Monte Carlo simulations and the multiple foil activation technique. The high energy neutron beams produced via the ${}^2\text{H}(\text{d},\text{n}){}^3\text{He}$ and ${}^3\text{H}(\text{d},\text{n}){}^4\text{He}$ reactions, have been extensively used for threshold reaction cross section measurements on various isotopes with the activation technique, while the low energy neutron beams from the ${}^3\text{H}(\text{p},\text{n}){}^3\text{He}$ reaction have been used for in-beam fission cross section measurements on actinides with Micromegas detectors.

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The details of the description and the characteristics of the neutron production facility will be presented in this paper along with an overview of the neutron activation campaign.

2 The neutron facility

The neutron facility at the tandem accelerator of NCSR “Demokritos” can deliver quasi-monoenergetic neutron beams at different energies through the following four reactions:

- $\sim 120\text{--}650$ keV via the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction
- $\sim 2\text{--}5.3$ MeV via the ${}^3\text{H}(p,n){}^3\text{He}$ reaction
- $\sim 4.0\text{--}11.4$ MeV via the ${}^2\text{H}(d,n){}^3\text{He}$ reaction
- $\sim 16\text{--}20$ MeV via the ${}^3\text{H}(d,n){}^4\text{He}$ reaction

The target assemblies for all these reactions are air cooled during the proton or deuteron beam irradiations. Two collimators of 5 and 6 mm diameter are used and the beam current is measured both at the collimators and the target and is kept at $\sim 700\text{nA}\text{--}1\ \mu\text{A}$. The tandem accelerator is currently under major upgrade through the program CALIBRA/EYIE (see Acknowledgements) and the beam current is expected to be considerably increased in the near future. The flux variation of the neutron beam is monitored by a BF₃ detector placed at a distance of 2–3 m from the neutron producing target. The spectra of the BF₃ monitor are stored at regular time intervals (~ 100 sec) in a separate ADC during the irradiation process, while the absolute flux of the beam is obtained with respect to the cross section of reference reactions (${}^{27}\text{Al}(n,\alpha)$, ${}^{197}\text{Au}(n,2n)$ and ${}^{93}\text{Nb}(n,2n)$) and varies from $10^5\text{--}10^6$ n/cm²s. The beam current on the target is also recorded in another ADC during the same time intervals, in order to test the reliability of the BF₃ counter during long irradiation periods. In addition, a BC501A scintillator detector is used for the investigation of the neutron beam energy distribution.

A neutron source is considered mono-energetic when the energy spectrum consists of a single line with an energy width which is much less than the energy itself. Such mono-energetic neutrons can be produced by two-body reactions, however a real source will not only produce these primary neutrons, but also “parasitic” ones, which vary in energy from the thermal region up to a few MeVs. Parasitic neutrons mainly stem from a) deuteron break up reactions, such as ${}^3\text{H}(d,pn){}^3\text{H}$, ${}^2\text{H}(d,np){}^2\text{H}$, which start to contribute significantly above 3.7 MeV deuteron energy b) from reactions of the d and p beam with the target material, as for example Mo, Ti and Cu and reactions with C, due to the carbon built up in the target and the beam line and even with O due to oxidization processes c) moreover, the deuteron beam that is implanted in the target yields background neutrons from the ${}^2\text{H}(d,n){}^3\text{He}$ reaction. In the case of gas targets the contribution from these background neutrons can relatively easily be determined via gas-in/gas-out measurements, for solid targets however, the task to perform a blank measurement using a non-tritium containing dummy target is a somewhat more demanding task d) finally, another contribution to the parasitic neutrons is coming from scattering of neutrons from the walls, ceiling and objects in the experimental room and the experimental set up, leading to a low energy tail in the neutron spectrum.

The study of neutron energy spectra generated by all the four above mentioned reactions, is carried out with detailed Monte Carlo simulations using the MCNP5 code [2]. The description of the neutron source imported in MCNP5 is generated by the NeuS-Desc (Neutron Source Description) code [3]. The program takes into consideration the energy loss, energy spread and angular straggling of the protons or deuterons in the target

assembly through the program SRIM 2008 and calculates average neutron energies, fluences and resolutions. The details of the neutron beams produced via these four reactions along with results from different techniques used for the neutron beam characterisation, will be presented below.

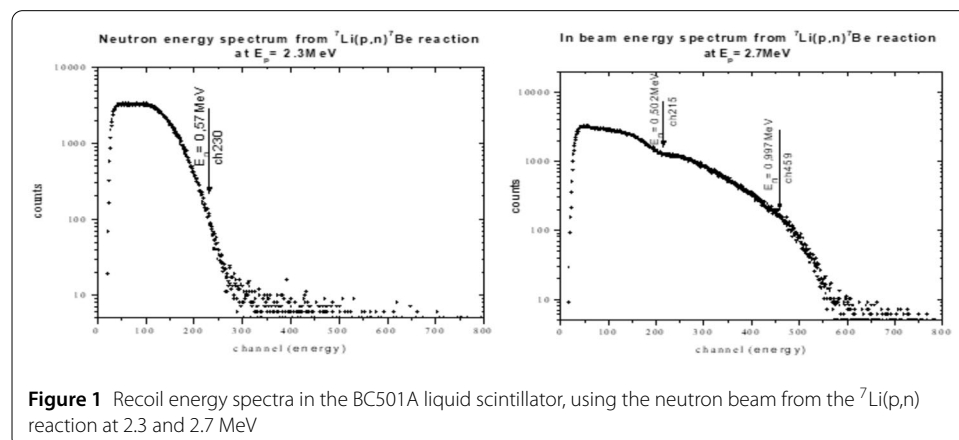
2.1 The ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction

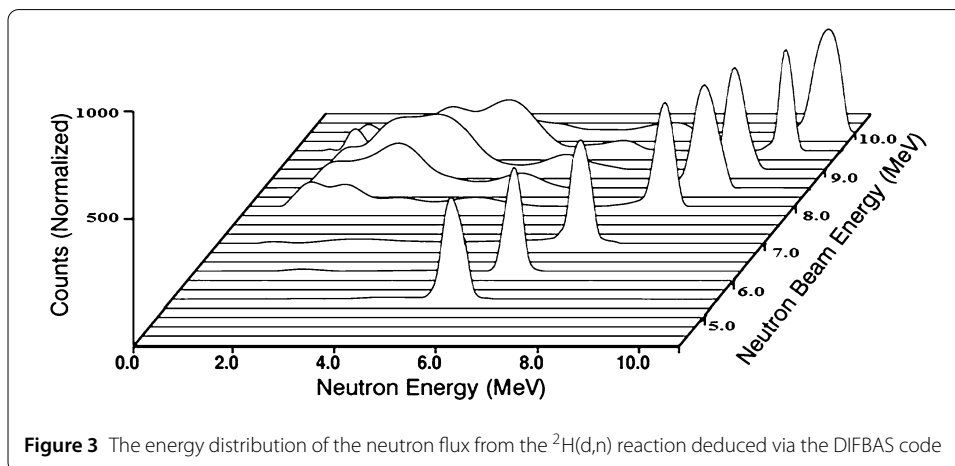
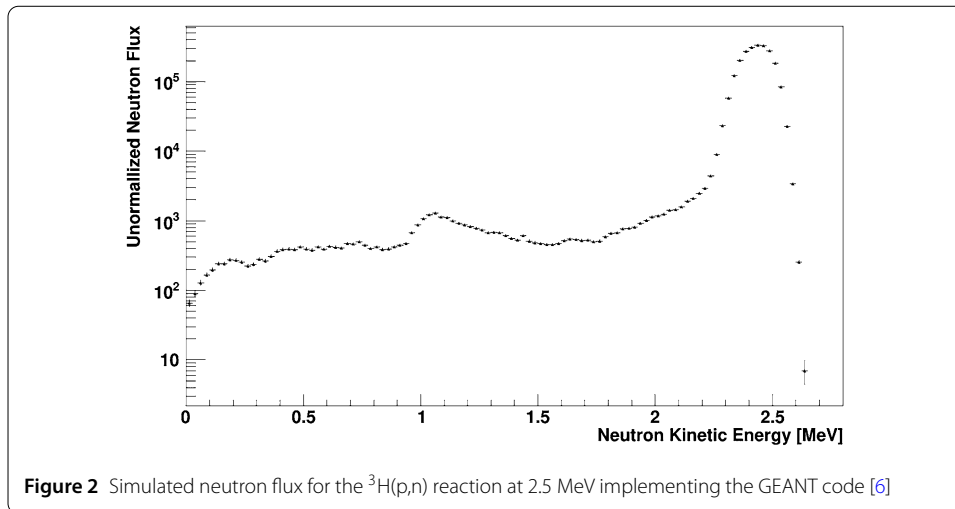
For the neutron production via the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction, a ${}^7\text{LiF}$ target on Al backing is used along with proton beams in the energy range 1.9–2.4 MeV to produce purely mono-energetic neutrons at zero degrees between 120 keV and 650 keV, respectively. At proton energies above 2.4 MeV, neutron emission to the 1st excited state in ${}^7\text{Be}$ at 429 keV is possible and produces a second group of mono-energetic neutrons. The BC501A scintillator detector was used to investigate the neutron energy spectra from this reaction. By rejecting the gamma pulses, the recoil energy spectra in the liquid scintillator can be reconstructed, as shown in Fig. 1 in the case of the ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction at proton beam energies set at 2.3 and 2.7 MeV, respectively. At 2.3 MeV, the recoil spectrum corresponds to purely mono-energetic neutrons of 0.57 MeV, as indicated by the arrow, while at 2.7 MeV, apart from the main production of neutrons at 0.997 MeV, a second bump appears in the spectrum corresponding to the 0.502 MeV neutrons arising from the first excited state of ${}^7\text{Be}$.

The spectra shown in Fig. 1 were used for the energy calibration of the BC501A scintillator detector in the low energy region.

2.2 The ${}^3\text{H}(p,n){}^3\text{He}$ reaction

For the neutron production via the ${}^3\text{H}(p,n){}^3\text{He}$ reaction, a Ti-tritiated target is used, consisting of a 2.1 mg/cm^2 Ti-T layer on a 1 mm thick Cu backing for good heat conduction. Two $5\text{ }\mu\text{m}$ Mo foils were placed in front of the target in order to degrade the beam energy. Proton beams in the energy range 3.4–6.5 MeV have been used to produce neutrons between 2 and 5.3 MeV. Purely mono-energetic neutrons are generated in the range $\sim 2\text{--}3$ MeV, while at higher energies (p,n) reactions on Mo, Ti, Cu, C and ${}^2\text{H}$ implanted in the TiT target from previous irradiations, become important and can lead to distinct peaks in the neutron spectra [4]. Extensive MCNP5 simulations coupled with NeuSDesc results, have been performed, as described in [4]. In an attempt to investigate the parasitic neutrons in more detail and include all the interactions of the proton beam with the experimental setup used to generate neutrons, biased Monte Carlo simulations have





been performed implementing the GEANT4 code [5]. Preliminary results for the 3.8 MeV proton beam leading to 2.5 MeV neutrons, are presented in Fig. 2.

2.3 The ${}^2\text{H}(d,n){}^3\text{He}$ reaction

For the neutron production via the ${}^2\text{H}(d,n){}^3\text{He}$ reaction, a 3.7 cm long and 1 cm diameter deuterium gas cell pressurized to ~ 1 atm is used, with a $5\ \mu\text{m}$ Mo entrance foil and a 1 mm Pt foil for the beam stop. The neutrons lie in the energy range ~ 4 –11.2 MeV, at the corresponding deuteron beam energies 0.8–8.2 MeV. The energy spectrum and the characteristics of the beam have been studied by means of the multiple foil activation technique in combination with the SULSA code [7] as well as with the liquid scintillator BC501A and deconvolution of its recoil energy spectra using the DIFBAS code [8]. The results from this investigation at neutron beam energies 5.5, 6.3, 7.0, 8.0, 8.5, 9.0, 9.5 and 10 MeV are described in detail in [9] and are shown in a three dimensional plot in Fig. 3. In this figure, the y-axis represents the energy of the main neutron beam, while the x-axis represents the neutron energies down to low energies to account for the “parasitic” neutrons which accompany the main beam. The neutron beams at 5.5 and 6.3 MeV are seen to be monoenergetic, while above 7 MeV “parasitic” low energy neutrons start to appear at energies around 4 MeV and become more and more significant as the main

neutron beam energy increases. These results are in consistency with the neutron energy spectra deduced with the code SULSA, within the limitations of the multiple foil activation technique. In addition, MCNP5 simulations have been performed in order to study the neutron flux energy distribution around 0° with respect to the beam line axis.

2.4 The $^3\text{H}(d,n)^4\text{He}$ reaction

For the neutron production via the $^3\text{H}(d,n)^4\text{He}$ reaction, the same Ti-tritiated target used in the case of the $^3\text{H}(p,n)^3\text{He}$ reaction, is implemented. The produced neutrons lie in the region 15.3–20.9 MeV at the corresponding deuteron beam energies 2.0–4.3 MeV. The study of neutron energy spectra generated by deuterons on the Ti-tritiated target was carried out utilizing the NeuSDesc (Neutron Source Description) output file for Monte Carlo simulations using the MCNP5 code. Furthermore, several foils were irradiated at different energies, most of them corresponding to reference reactions, in order to extract experimentally the neutron beam fluence [10]. By taking into account the detailed geometry of the experimental setup for each irradiation, the neutron fluence in the successive foils was simulated and the results were compared with the experimental ones. As an example, the results at (17.9 ± 0.3) MeV for the irradiation of Al–Au–Ir–Al–Er–Al–Au sequence of foils in the target assembly [11], are presented in Fig. 4. The experimental and simulated neutron fluence in the successive foils seem to be in very good agreement, indicating that the simulations are successful and can thus be trusted for the estimation of the neutron fluence.

The simulated neutron fluence energy spectrum at 17.9 MeV is shown in Fig. 5.

The long tail of parasitic neutrons is illustrated in Fig. 5, which is about three orders of magnitude lower than the main beam. However, these neutrons may cause significant problems in cross section measurements of reactions which are sensitive to low energy neutrons, such as fission reactions. Thus, in facilities where the neutron ToF technique is not applied, only reactions with a high energy threshold can be safely measured with mono-energetic neutrons, since the low energy parasitic neutrons cannot affect the cross section measurements, provided that the reference reaction used for the determination of the absolute neutron flux also has a threshold in the same energy region.

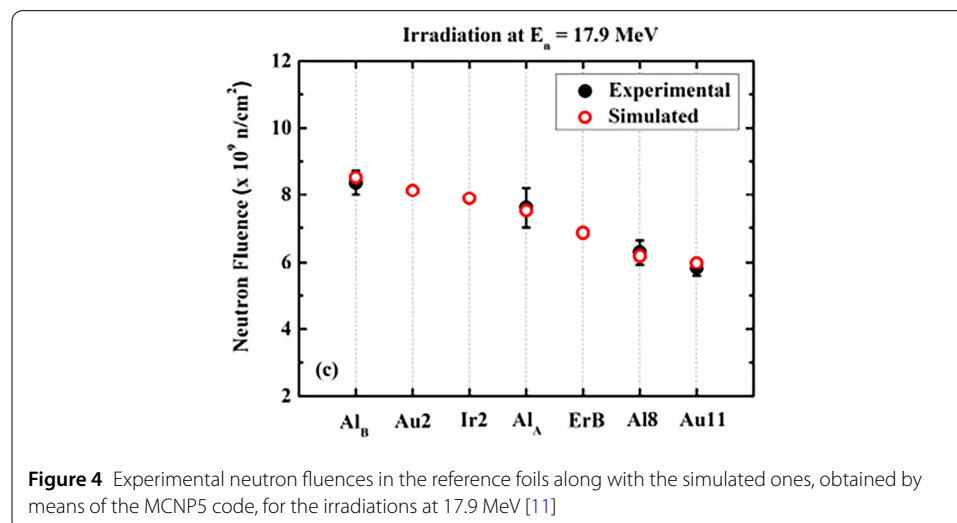


Figure 4 Experimental neutron fluences in the reference foils along with the simulated ones, obtained by means of the MCNP5 code, for the irradiations at 17.9 MeV [11]

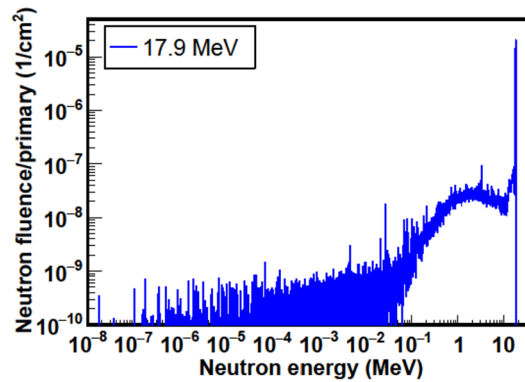


Figure 5 Simulated neutron fluence from MCNP5 at 17.9 MeV

3 Activation cross section measurements

3.1 Experimental procedure

The neutron beam at NCSR “Demokritos” has been extensively used over the past 15 years by the NTUA (National Technical University of Athens) group, for the measurement of (n,2n) and occasionally (n,3n), (n,p), (n, α) reaction cross sections on several isotopes of Am [12–14], Hf [15, 16], Ge [17, 18], Ir [19, 20] and Au [21, 22], with the activation technique. For all these measurements neutron beams were used in the energy region from ~ 7 –11 MeV produced via the $^2\text{H}(d,n)^3\text{He}$ reaction as well as from ~ 15 –21 MeV derived via the $^3\text{H}(d,n)^4\text{He}$ reaction. High purity foils were used for these measurements and were stacked between two Al reference foils for the irradiation. Apart from the $^{27}\text{Al}(n,\alpha)$ reference reaction other reactions such as $^{197}\text{Au}(n,2n)$ and $^{93}\text{Nb}(n,2n)$ have been also used to accurately determine the neutron flux. After the end of the irradiation the induced γ -ray activity of the samples and the reference targets were measured off-line by HPGe detectors. The activity measurements of all samples were normally carried out at a distance of ~ 10 cm from the detector window, thus there was no need for significant pile-up or true coincidence summing effect corrections. At the same distance, a ^{152}Eu point source was placed in order to determine the absolute efficiency of each detector. Additionally, the experimental set up was always simulated with the use of the MCNP code, for the estimation of the neutron flux, the self-absorption of the γ -rays in the sample, the effect of parasitic neutrons which accompany the beam etc.

3.2 Theoretical calculations

All the above mentioned reactions are relevant to practical applications for nuclear technology, high energy neutron dosimetry, detector physics, medical applications etc., but they are also important for the investigation of model parameters in statistical model calculations. Thus, the study of the measured reactions is usually accompanied by theoretical cross section calculations in the framework of the Hauser–Feshbach theory, using the latest versions of the EMPIRE [23] and TALYS [24] codes. The calculations are carried out in a wide energy range for the measured data of each reaction as well as for the data reported in literature.

From all the measured neutron induced reaction cross sections on isotopes of Am, Hf, Ir, Ge and Au, the reactions on Au and Ir will be mentioned in more detail, since the (n,2n) reactions on ^{191}Ir and ^{197}Au lead to the population of both ground and isomeric states of

the residual nuclei. The theoretical study of these reactions, due to the existence of high spin isomeric states, is a powerful tool for obtaining information on the structure of the involved nuclei and thus constitutes an open field of study. The simultaneous reproduction of the isomeric and ground state cross sections along with other channels where data are available in literature, sets a significant constraint, rendering theoretical calculations quite sensitive to the choice of specific nuclear model parameters.

In the case of the $^{197}\text{Au}(n,2n)^{196}\text{Au}^{g+m_1+m_2}$ and $^{197}\text{Au}(n,2n)^{196}\text{Au}^{m_2}$ reactions, cross sections were measured in the energy range 9.0–10.5 MeV [21] and 15.3–20.9 MeV [22]. This data, along with all other data available in literature, as well as data for the (n,3n), (n,elastic), (n, α), (n,p) and (n,total) competing reactions, were simultaneously reproduced in a satisfactory way by both EMPIRE and TALYS calculations [22]. The high angular momenta treatment in the EGSM level density model in the EMPIRE code, helped to reproduce the experimental values of the isomeric cross sections as it affects more efficiently the spin distribution of level densities above the critical excitation energy. In TALYS, a small increase of the spin cut-off parameter via the “Rspincut” keyword, was sufficient to successfully reproduce the cross section of the isomeric state along with all other competing channels.

In the case of the $^{191}\text{Ir}(n,2n)^{190}\text{Ir}^{g+m_1}$ and $^{191}\text{Ir}(n,2n)^{190}\text{Ir}^{m_2}$ reactions, cross sections were measured in the energy range 10–11.3 MeV [19] and 15.3–20.9 MeV [20]. This data, along with all other data from literature, as well as data for the (n,3n), (n,p) and (n,total) competing reactions and data for the other isotope $^{193}\text{Ir}(n,2n)$, $^{193}\text{Ir}(n,\alpha)$, $^{193}\text{Ir}(n,p)$ and $^{193}\text{Ir}(n,\text{total})$ were reproduced in a satisfactory way by the EMPIRE and TALYS calculations [20]. It is interesting to note that these calculations were performed with a similar combination of model parameters that had also been successfully used in the case of the neighbouring ^{197}Au nucleus. This constitutes an encouraging confirmation of how accurately the theoretical models can reproduce the experimental results in this mass region. These similarities in the theoretical parametrization are not that surprising since both Au and Ir nuclei belong to the transitional region from well deformed to spherical nuclei near the shell closure $Z = 82$ (Os–Pb region), where high spin intruder configurations result in high spin isomeric states unable to communicate with neighboring states. It would be quite interesting to continue this investigation for the next nucleus Tl in this mass region. Indeed, measurements are planned to be carried out in the near future for the $^{203}\text{Tl}(n,2n)$ reaction at the neutron facility of NCSR “Demokritos”.

4 Summary

Neutron beams are produced at the tandem accelerator of NCSR “Demokritos” in Athens at energies varying in the range from about 120 keV to 21 MeV by means of the following proton and deuteron induced reactions: $^7\text{Li}(p,n)^7\text{Be}$, $^3\text{H}(p,n)^3\text{He}$, $^2\text{H}(d,n)^3\text{He}$ and $^3\text{H}(d,n)^4\text{He}$. A comprehensive understanding of the energy dependence of the produced neutron beam flux is of major importance for the reliability of neutron induced reaction cross section measurements. Thus, for all these four reactions several techniques have been used for this investigation, such as the unfolding method applied to the multiple foil activation results and the deconvolution of recoil energy spectra taken via the BC501A liquid scintillator detector at various neutron energies. Furthermore, extensive Monte Carlo simulations by means of the MCNP5 and GEANT4 codes have been performed to study the neutron energy spectra in conjunction with other experimental techniques like gas-in

gas-out tests, irradiations with and without a Cd foil in front of reference foils etc. The neutron beams have been used for the measurements of $(n,2n)$ and occasionally $(n,3n)$, (n,p) , (n,α) reaction cross sections on isotopes of Am, Hf, Ir, Ge and Au, with the activation method. The investigation of these reaction cross sections is followed by theoretical calculations implementing the codes EMPIRE and TALYS. The tandem accelerator at NCSR “Demokritos” is currently under renovation and in the near future these techniques will be applied again for the investigation of the upgraded neutron beams, while the neutron activation measurements will continue to be an important research project of the Nuclear Physics group at the National Technical University of Athens.

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Availability of data and materials

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Declarations

Competing interests

The author declares that they have no competing interests.

Author contributions

The author wrote, read and approved the final manuscript.

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