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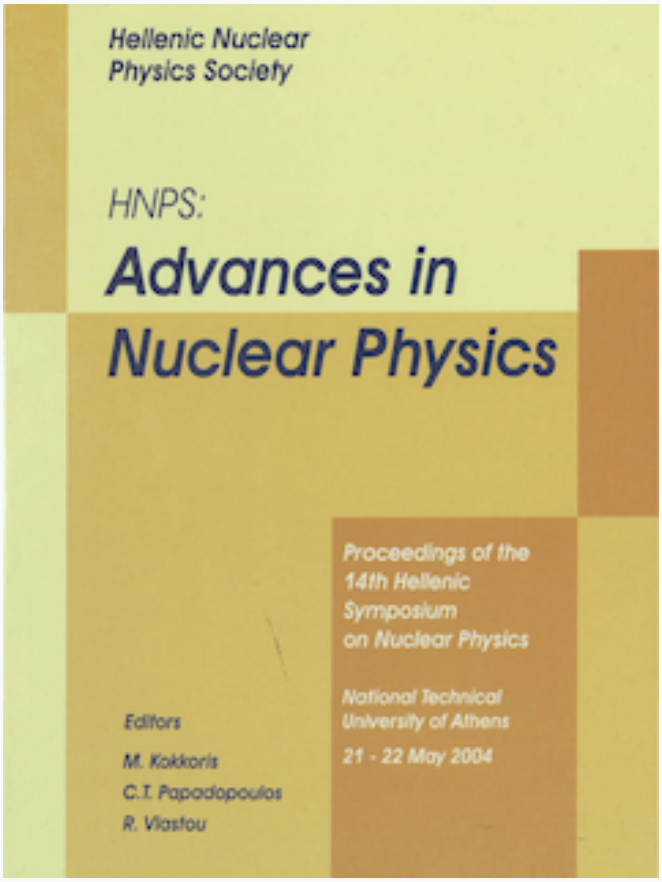


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# The Neutron Facility at NCSR "Demokritos"- Implementation in the Case of the $^{232}$Th(n,2n) and $^{241}$Am(n,2n) Reactions

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**Abstract**

In the 5.5 MV tandem T11/25 Accelerator Laboratory of NCSR "Demokritos" monoenergetic neutron beams can be produced in the energy ranges 120-650 keV, 4-11.5 MeV and 16-20.5 MeV by using the $^{7}$Li(p,n), $^{2}$H(d,n) and $^{3}$H(d,n) reactions, respectively. The corresponding beam energies and ions delivered by the accelerator, are 1.92-2.37 MeV protons, 0.8-9.6 MeV deuterons and 0.8-3.7 MeV deuterons, for the three reactions, respectively. Experimental results for neutron energies from threshold up to 11.5 MeV and at 17.1 MeV will be given for the $^{232}$Th(n,2n)$^{231}$Th reaction, while for the $^{241}$Am(n,2n)$^{240}$Am reaction, preliminary cross section data at 10.4, 10.6 and 17.1 MeV will be discussed. In the framework of the CERN n-TOF collaboration, the cross section of these reactions have been measured relative to the $^{197}$Au(n,2n)$^{196}$Au, $^{27}$Al(n,$\alpha$)$^{24}$Na and $^{93}$Nb(n,2n) reaction cross sections, by using the activation method. In addition to the experimental work, theoretical Statistical model calculations are being carried out using the computer code STAPRE/F. The results are compared to the experimental data.

## 1 Introduction

The study of neutron induced reactions has become recently of particular interest, not only for their importance to fundamental research in Nuclear Physics and Astrophysics, but also for practical applications in nuclear technology, medicine and industry. The main technological applications are related to the

design of innovative Accelerator Driven Systems (ADS) for the future production of clean and safe nuclear energy as well as for the incineration of nuclear waste [1-6]. These tasks require knowledge of complete and precise cross sections for neutron (n,f), (n,$\gamma$) and (n,xn) reactions on a variety of isotopes. Capture and fission data are needed for the minor actinides, capture cross sections for the main fission products and (n,xn) reactions for transuranic isotopes as well as for structural and coolant materials. The available compilation data-bases, based on both experimental and theoretical evaluations, present many differences and discrepancies [7], due to different theoretical models and fitting procedures that are used. So, they cannot be considered as reliable basis for planning in detail these Accelerator Driven Systems [8, 9].

In order to produce accurate and consistent neutron cross section data, a neutron time of flight facility has been set in operation at CERN [10], as a large international collaboration under the name n-TOF. It is based on the spallation mechanism for neutron production, of a lead target bombarded by 20 GeV protons from the PS accelerator at CERN. High intensity neutron beams are generated, in the energy interval from 1eV to 250 MeV and the determination of their energy is reconstructed by the measured time of flight [11]. Complementary cross section measurements with monoenergetic neutron beams are, however, essential for testing the reliability of the results. The n_TOF measurements will provide a complete and reliable database over a wide energy range for nuclear energy research. They will also allow verification of nuclear reaction models and improve the evaluated neutron reaction database.

In view of the above remarks, the neutron facility at the 5.5MV tandem T11/25 Accelerator of NCSR "Demokritos", has been used for (n,2n) cross section measurements with monoenergetic neutron beams. The facility has been upgraded and extended to cover broader energy range. The recent developments and techniques used for the production of monoenergetic neutron beams at "Demokritos", will be presented in this paper. In the context of the n-TOF collaboration at CERN, the $^{232}$Th(n,2n) reaction has been measured by using the activation technique, in the energy range 7.5-11.5 MeV [12], as well as at 17.1 MeV, and the $^{241}$Am(n,2n)$^{240}$Am reaction at 10.4, 10.6 and 17.1 MeV. The experimental results and the theoretical investigation of the former reaction will also be described in the present report.

## 2 The Neutron Facility

The production of monoenergetic neutron beams at the 5.5MV tandem T11/25 Accelerator at the NCSR "Demokritos" can be achieved by using different reactions for different energy regions, as described in [13].

For low energies between 120 and 650 keV neutrons can be produced via the $^{7}$Li(p,n)$^{7}$Be reaction by using a $^{7}$LiF target on Al backing and a proton beam in the energy range 1.9-2.4 MeV, where the neutrons produced at zero degrees are monoenergetic. At higher proton energies, up to 5.5MeV, the zero degree neutrons can reach 3.8 MeV energy, but they are not strictly monenergetic since they contain a $\sim$10% contribution of neutrons coming from the first excited state of $^{7}$Be. The neutron beam flux in all cases is of the order of $10^5$ n/cm$^2$sec for a proton beam current $\sim$1 $\mu$A.

For the middle energies between 4.0 and 11.5 MeV, neutrons are produced via the $^{2}$H(d,n)$^{3}$He reaction by using deuteron beam in the energy range 0.8-9.6 MeV and a gas cell target 3.7 cm long made of stainless steel, as described in Ref.[14]. The entrance window is a 5$\mu$m Mo foil and the beam stops on a 1mm Pt foil. The deuterium gas pressure can be monitored and refilled electronically when the cell pressure falls below a preset level. For deuteron currents up to 5$\mu$A and deuterium gas pressures up to 1.5 atm, the neutron flux at $0^o$ is of the order of $10^6$ n/cm$^2$sec.

For the higher energies between 16.0 and 20.5 MeV, neutrons are produced via the $^{3}$H(d,n)$^{4}$He reaction by using deuteron beam in the energy range 0.8-3.7 MeV and a Ti tritiated target of 5Ci activity on a Ag backing, for good heat conduction. To avoid heating and out gazing, the target is water-cooled during the irradiation with the deuteron beam of $\sim$2$\mu$A. The corresponding neutron beam flux at $0^o$ is of the order of $10^5$ n/cm$^2$sec.

Neutron energies are calculated from reaction kinematics by taking into account the proton or deuteron energy loss in the target, the cross section of the reaction and the irradiation geometry [15]. Lower energies of the neutron beam can be achieved by placing the target for the reaction under investigation at an angle to the neutron beam, leading to lower flux and lower beam energy resolution. In all three cases the flux variation of the neutron beam is monitored by using a $BF_3$ detector whose spectra are stored at regular time intervals in a separate ADC during the irradiation process. In addition, the beam current on the target is also recorded at the same time intervals in another ADC, in order to test the reliability of the $BF_3$ counter during the long irradiation time. The absolute flux of the beam can be obtained with respect to reference reactions, such as $^{197}$Au(n,2n),$^{27}$Al(n,$\alpha$) and $^{93}$Nb(n,2n), whose cross sections are well determined in the literature.

## 3 The $^{232}$Th(n,2n) and $^{241}$Am(n,2n) reactions

In the frame of the n-TOF collaboration, the cross section of the $^{232}$Th(n,2n) reaction, which is important for the Th-U cycle, has been measured in the

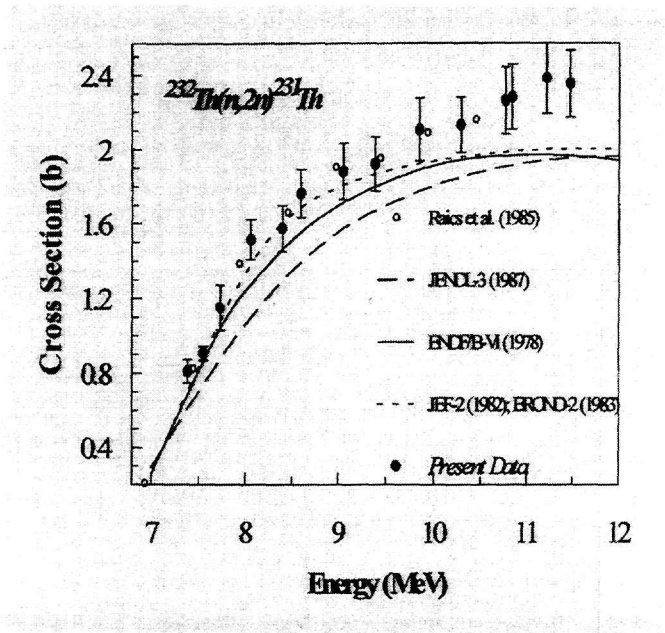


Fig. 1. Experimental cross sections of the $^{232}$Th(n,2n) reaction in comparison with existing data from Ref. [16] and evaluations from different databases [7].

energy range 7.5-11.5 and 17.1 MeV by using the activation method. The $\beta$-decay of the produced nucleus $^{231}$Th leads to the excited states of $^{231}$Pa, with a half-life of 25.52 h. The deexcitation of $^{231}$Pa proceeds via gamma rays, the most prominent of which for the activation measurement, is the 25.7 keV transition. Induced gamma activity was measured with an 80 mm$^2$ Si(Li) detector, continuously for a few days after the exposure to the neutron beam. The experimental details about these measurements and the analysis of the data, are described in Ref. [12].

The cross section values extracted by the experimental data are presented in Fig. 1 together with the most recent data by Raics et al. [16] and the evaluations from four reference libraries of neutron data. In the overlapping energy region the data of Raics et al. seem to coincide with the results presented here, while the evaluated values from the various databases [7], seem to underestimate the experimental ones at energies above ~10 MeV.

The $^{241}$Am(n,2n)$^{240}$Am reaction is of particular importance for the design of ADS, since the production of an extra neutron by the reaction affects the neutron balance in the core of the reactor and Americium is highly radiotoxic and abundant isotope among the actinides in the spent nuclear fuel. The cross section of the reaction has been measured at 10.4 and 10.6 MeV, via

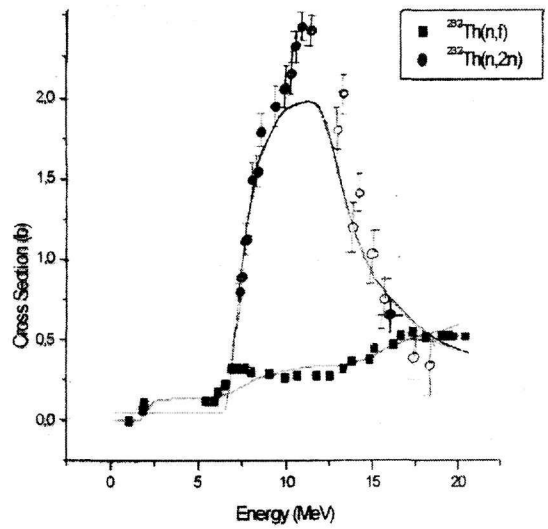


Fig. 2. Experimental cross sections of the $^{232}$Th(n,2n) and $^{232}$Th(n,f) reactions in comparison with statistical model calculations. The data for the $^{232}$Th(n,f) cross sections were taken from the EXFOR experimental nuclear reaction database. The solid circles represent the data of the present work, while the open circles were taken from literature.

the $^{2}$H(d,n) reaction and at 17.1 MeV, via the $^{3}$H(d,n) reaction, by using the activation method. The target is $Am_2O_3$ of 37GBq activity, enclosed in a lead cylindrical box of 3mm thickness to ensure almost complete attenuation of the 59.6keV $\gamma$-ray from the decay of $^{241}$Am, both for radioprotection reasons and for reducing the detector dead time during the measurements. The reaction leads to the formation of $^{240}$Am in the relatively short-lived ($\tau$ = 50.8h ) state, which decays by electron capture to $^{240}$Pu, whose deexcitation contains two strong transitions 987.8 and 888.8 keV of absolute intensity 73.2% and 25.1%, respectively. These characteristic $\gamma$-rays of the $^{241}$Am(n,2n)$^{240}$Am reaction can be used for the determination of the cross section, by applying the activation method. Indeed, after continuous irradiation for ~100h, the induced activity of the target was measured by using HPGe detectors of relative efficiency 9% and 50% . The experimental details about these measurements and the analysis of the data, are described in the next presentation in this conference.

## 4 Statistical model calculations

The excitation function for the cross sections of the $^{232}$Th(n,2n) and fission reactions have been calculated in the framework of the Hauser-Feshbach theory using a modified version of the code STAPRE/F [17]. The code has been de-

signed to estimate energy-averaged cross sections for particle-induced reactions with several emitted particles and gamma-rays under the assumption of sequential evaporation. Discrete energy levels, branching ratios and transmission coefficients, required as input parameters in the code, were taken from [18]. Fissioning nuclei level densities at fission saddle deformations were extracted by the generalized superfluid model [18-20]. The values of the parameters used in these calculations are summarized in Table 1.

Table 1
Parameters used for the statistical model calculations

| Nucleus | $\alpha$ ($MeV^{-1}$) | $\Delta$ (MeV) | Fission Barrier Height (MeV) | | Fission Barrier Curvature (MeV) | | $\Delta$(MeV) For fissioning nuclei |
|---|---|---|---|---|---|---|---|
| | | | Inner Saddle | Outer Saddle | Inner Saddle | Outer Saddle | |
| $^{233}$Th | 24.4 | 0.786 | 5.1 | 6.65 | 0.7 | 0.5 | 0.806 |
| $^{232}$Th | 24.5 | 0.788 | 5.8 | 6.7 | 0.9 | 0.6 | 0.806 |
| $^{231}$Th | 25.5 | 0.800 | 6.0 | 6.7 | 0.7 | 0.5 | 0.830 |
| $^{230}$Th | 27.6 | 0.791 | | | | | |
| | | | | | | | |

Preliminary results of the statistical model calculations for the $^{232}$Th(n,2n) and $^{232}$Th(n,f) reactions are compared to the data in Fig. 2 and in general are seen to reproduce the overall trend of the excitation function.

They seem however, to underestimate the observed (n,2n) cross section in the energy region of 10 MeV. Furthermore, the fission cross section which increases around 7 MeV, can not be reproduced by the calculations, which are still under progress.

## 5 Summary

The neutron facility at the 5.5 MV tandem T11/25 Accelerator of NCSR "Demokritos" can deliver monoenergetic neutron beams in the energy range 120-650 keV, 4-11.5 MeV and 16-20.5 MeV via the $^{7}$Li(p,n), $^{2}$H(d,n) and $^{3}$H(d,n) reactions, respectively, at a maximum flux of the order of $10^{5}$-$10^{6}$ n/cm$^{2}$sec.

On the context of the CERN n-TOF collaboration, the cross section of the $^{232}$Th(n,2n)$^{231}$Th reaction has been measured between 7.5 - 11.5 and at 17.1

MeV, relative to the $^{27}Al(n,\alpha)^{24}Na$ reaction cross sections, by using the activation method. In addition, cross section measurements of the $^{241}Am(n,2n)$ reaction were carried out at energies 10.4, 10.6 and 17.1 MeV, relative to the $^{27}Al(n,\alpha)^{24}Na$, $^{197}Au(n,2n)^{196}Au$ and $^{93}Nb(n,2n)^{92}Nb$ reaction cross sections. Preliminary theoretical statistical model calculations are also presented for the $^{232}Th(n,2n)^{231}Th$ reaction, while for the $^{241}Am(n,2n)^{240}Am$ data, theoretical calculations are in progress. The results have been compared to the experimental data for the $^{232}Th(n,2n)^{231}Th$ and seem to be in reasonable agreement.

## Acknowledgments

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