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Neutron induced reactions at the Athens Tandem accelerator NCSR “Demokritos”

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Abstract

The neutron facility at the 5.5 MV Tandem T11/25 accelerator of NCSR “Demokritos” has been used to produce monoenergetic neutron beams in the energy range 120-650 keV and 4-11.5 MeV via the ${}^7\text{Li}(p,n)$ and ${}^2\text{H}(d,n)$ reactions, respectively. The flux variation of the neutron beam was monitored by using a BF_3 detector, while an investigation of the energy dependence of the neutron fluence has been carried out with a liquid scintillator BC501A detector as well as with the multiple foil activation technique. The ${}^{232}\text{Th}(n,2n){}^{231}\text{Th}$ and ${}^{241}\text{Am}(n,2n){}^{240}\text{Am}$ as well as $(n,2n)$, (n,p) and (n,α) reactions on natural Ge and Hf isotopes, have been investigated from threshold up to 11.5 MeV, by using the activation method.

1 INTRODUCTION

The design of innovative Accelerator Driven Systems (ADS) for incineration of nuclear waste and energy generation, requires complete knowledge of cross sections for neutron induced processes [1,2]. Studies of neutron induced reactions are also of considerable significance to fundamental research in Nuclear Physics [3–5] and Astrophysics. The available compilation data-bases, based on both experimental and theoretical evaluations, present many differences and discrepancies, due to different theoretical models and fitting procedures that are used. So, they cannot be considered as reliable basis for practical applications and for testing nuclear models [6].

In view of these remarks, the neutron facility at the 5.5MV Tandem T11/25 accelerator of NCSR “Demokritos”, has been used for the measurement of $(n,2n)$, (n,p) and (n,α) threshold reaction cross sections induced by monoenergetic neutron beams. In addition, an investigation of the neutron beam

characteristics has been performed by means of a new BC501A liquid scintillator detector as well as by using the multiple foil activation method. The preliminary results of this investigation will be presented in this work.

2 EXPERIMENTAL METHOD

In the 5.5 MV Tandem T11/25 accelerator Laboratory of NCSR "Demokritos" monoenergetic neutron beams have been produced in the energy ranges 120-650 keV and 4-11.5 MeV by using the ${}^7\text{Li}(p,n)$ and ${}^2\text{H}(d,n)$ reactions and subsequently used for neutron induced cross section measurements with the activation method [7]. The flux of the neutron beam is of the order of $10^5 - 10^6 n/(cm^2 s)$ and its variation is monitored by a BF_3 detector, whose spectra are stored at regular time intervals (≈ 100 sec) in a separate ADC during the irradiation. In addition, the integrated current of the deuteron beam on the target is also recorded for the same time intervals in another multi-scaling ADC, in order to test the reliability of the BF_3 counter during long irradiation periods. Furthermore, a new BC501A liquid scintillator detector has been recently used for the investigation of the energy distribution of the neutron beam. The absolute flux of the beam can be obtained with respect to reference reactions, such as ${}^{197}\text{Au}(n,2n)$, ${}^{27}\text{Al}(n,\alpha)$ and ${}^{93}\text{Nb}(n,2n)$, whose cross sections are well determined in the literature. In most cases the same reference foils are placed before and after the target and the mean value of the flux is considered to be the actual flux of the neutron beam. The experimental values were consistent with the simulated results performed with the Monte Carlo code MCNP [8].

Both reference and target samples are exposed to neutron beam and the induced activity of product radionuclides is measured with two HPGe detectors of 56% and 80% properly shielded with lead blocks to reduce the contribution of the natural radioactivity. The efficiency of the detectors at the position of the activity measurements is determined via a calibrated ${}^{152}\text{Eu}$ source. The cross section is determined by measuring the characteristic gamma rays from the decay of the residual nuclei, corrected for self absorption of the sample, coincidence summing effects of cascading gamma rays and counting geometry. The decay of product nuclide over the whole time range and the fluctuation of the neutron beam flux over the irradiation time are also taken into account.

3 STUDY OF NEUTRON BEAM ENERGY DISTRIBUTION

The BC501A scintillator detector was used for the investigation of the neutron beam energy distribution. The results from the pulse shape discrimination

technique, used to reject γ -ray induced pulses in the organic scintillator material, from neutron pulses, are shown in Fig. 1. In the case of neutron beam

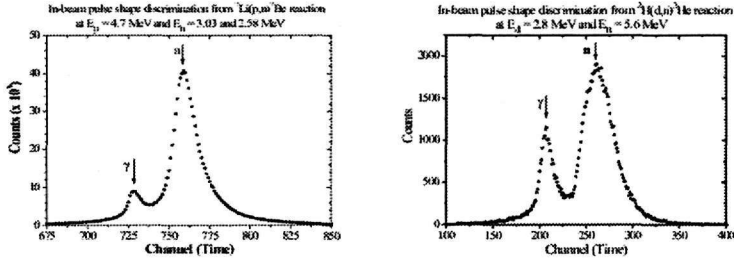


Fig. 1. Pulse shape discrimination pulses in the BC501A liquid scintillator, using neutron beam from the ${}^7\text{Li}(p,n){}^7\text{Be}$ and ${}^2\text{H}(d,n)$ reactions.

from the ${}^7\text{Li}(p,n)$ reaction, the discrimination between neutrons and gammas is quite good, despite of the fact that the neutrons are not purely monoenergetic, due to the production of ${}^7\text{Be}$ both at its ground and first excited state at this proton energy. In the case of neutron beams at higher energies produced by means of the ${}^2\text{H}(d,n)$ reaction, the discrimination between gammas and neutrons at 5.6 MeV shown in figure 1, is again quite good, while the production of gamma-rays is considerably enhanced, compared to the production of neutrons, as the energy increases. By rejecting the gamma pulses, the recoil

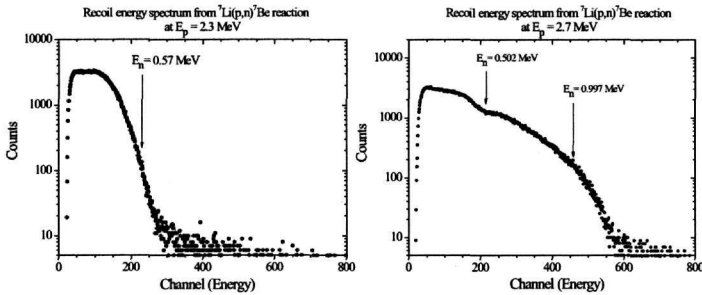


Fig. 2. Recoil energy spectrum in the BC501A liquid scintillator, using neutron beam from the ${}^7\text{Li}(p,n)$ reaction at two different proton energies.

energy spectra in the liquid scintillator can be produced, as shown in Fig. 2 for the cases of ${}^7\text{Li}(p,n){}^7\text{Be}$ reaction at proton energies 2.3 and 2.7 MeV, respectively. At 2.3 MeV, the recoil spectrum corresponds to purely monoenergetic 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 ${}^7\text{Be}$ at 0.429 MeV. The spectra shown in Fig. 2 have been used for the energy calibration of the BC501A scintillator detector

in the low energy region. The same process has been applied at higher neutron energies, produced by means of the $^2\text{H}(\text{d},\text{n})$ reaction, as shown in Fig. 3 for two deuteron energies 2.8 and 4.6 MeV, respectively, corresponding to neutron energies 5.6 and 7.6 MeV, indicated by arrows in the two spectra. In these two energies, additional spectra were taken with empty cell (gas out), in order to investigate the production of parasitic neutrons coming from the

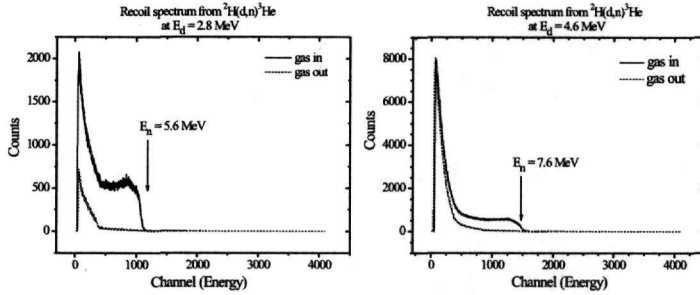


Fig. 3. Recoil energy spectrum in the BC501A liquid scintillator, using neutron beam from the $^2\text{H}(\text{d},\text{n})$ at two different deuteron energies as well as with and without deuterium gas in the cell.

cell itself. As can be observed in Fig. 3, there is a considerable production of low energy parasitic neutrons, especially in the high energy spectrum, mainly coming from the reaction of the deuteron beam with the Mo and Pt window foils of the cell. Furthermore, parasitic neutrons may result from the deuteron beam bombarding the collimators along with neutrons scattered by the walls of the laboratory or heavy masses around the target area.

In order to further investigate the neutron beam flux, the multiple foil activation method has been applied, which is widely used for the determination of the neutron flux density around the irradiated samples along with unfolding techniques [9]. Reactions of different thresholds $^{197}\text{Au}(\text{n},2\text{n})^{196}\text{Au}$, $^{27}\text{Al}(\text{n},\alpha)^{24}\text{Na}$, $^{56}\text{Fe}(\text{n},\text{p})^{56}\text{Mn}$, $^{115}\text{In}(\text{n},\text{n}')^{115\text{m}}\text{In}$ and $^{93}\text{Nb}(\text{n},2\text{n})^{92\text{m}}\text{Nb}$ have been used and the results are shown in table 1. The results are shown on table 1, which contains the information concerning the energy threshold of each reaction, the half lives of the residual nuclei along with the most prominent γ -rays resulting from their deexcitation, as well as the average value of neutron beam flux deduced from each reaction. High purity foils of ^{93}Nb , ^{197}Au , ^{115}In , ^{56}Fe and ^{27}Al for the reference reactions were placed in close contact at a distance of 9 cm from the neutron gas cell for the irradiation. Induced activities of product radionuclides were measured off-line by the 56% HPGe detector system and the neutron beam flux was deduced, by utilizing cross section values for each reaction from the IRDF-2002 compilation by IAEA [10].

The first three reactions in table 1 are characterized by high energy threshold, they practically open above 6 MeV, thus they are not influenced by the low energy parasitic neutrons involved in the neutron beam. As expected, the mean

Table 1

Nuclear data of reference reactions.

$E_n(\text{MeV})$	Reaction	$T_{1/2}$	$E_\gamma (\text{keV})$	$E_{thr}(\text{MeV})$	$f(10^6 \frac{n}{\text{cm}^2 \text{s}})$
9.7	$^{93}\text{Nb}(n,2n)^{92m}\text{Nb}$	10.5 d	934.5	9	0.9 ± 0.1
9.7	$^{197}\text{Au}(n,2n)^{196g}\text{Au}$	6.2 d	335.6	8	1.1 ± 0.1
9.7	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	14.9 h	1368.6	6	1.1 ± 0.1
11.5	$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	14.9 h	1368.6	6	3.1 ± 0.1
11.5	$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	2.6 h	846.8	4.5	2.7 ± 0.1
11.5	$^{115}\text{In}(n,n')^{115m}\text{In}$	4.5 h	336.3	0.5	1.3 ± 0.1

values of the neutron flux f from the three reactions in table 1, are identical within their errors. On the contrary, the last three reactions, involve different energy thresholds, ranging from 0.5 to 6 MeV. The low threshold reactions can be populated by both the high energy as well as low energy parasitic neutrons which are characterized by different reaction cross sections. Thus the neutron flux values deduced by these low threshold reactions are contaminated by the parasitic neutrons, as indicated by the discrepancies in f values presented in table 1. From the three reactions included in this table, reliable results for the beam flux can only be achieved by the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reaction, which is not influenced by the low energy neutrons. It is thus concluded that only threshold reactions can be safely measured with monoenergetic neutrons, since the low energy parasitic neutrons involved in the beam, cannot affect the cross section measurements.

In order to determine the neutron flux energy distribution in the near future, the unfolding method will be applied to the multiple foil activation results, in corroboration with the results from deconvolution of recoil energy spectra taken with the BC501A liquid scintillator detector at various neutron energies.

4 REACTION MEASUREMENTS

Two areas of investigation of such threshold reactions have been performed at the tandem Accelerator Laboratory of NCSR "Demokritos". Reactions relevant to nuclear energy applications, $^{232}\text{Th}(n,2n)$ and $^{241}\text{Am}(n,2n)$, as well as $(n,2n)$, (n,p) , (n,α) reactions on natural Ge and Hf, by using the activation method. Within the frame of the CERN n-TOF collaboration, the $^{232}\text{Th}(n,2n)$ reaction which is important for the Th-U cycle leading to the highly radiotoxic ^{232}U , has been investigated in the energy range from 7.5-11.5 MeV. The experimental details about these measurements and the analysis of the data, are described in ref. [11]. In addition, the $^{241}\text{Am}(n,2n)$ reaction has been measured at four energies from 8.8 to 11.4 MeV. Am is one of the most abundant iso-

topes in spent nuclear fuel and one of the most highly radiotoxic among the actinides and the data available in the literature are limited at energies around 14.5 MeV. The experimental details about this reaction and the analysis of the data, are described in another contribution in this conference. Concerning natural Ge and Hf, among the possible reaction channels produced by the neutron irradiation, the only ones involving reasonable lifetimes and being able to be measured by the activation method, are the $^{72}\text{Ge}(n,p)^{72}\text{Ga}$, $^{72}\text{Ge}(n,\gamma)^{69m}\text{Zn}$, $^{73}\text{Ge}(n,p)^{73}\text{Ga}$, $^{74}\text{Ge}(n,\alpha)^{71m}\text{Zn}$, $^{174}\text{Hf}(n,2n)^{173}\text{Zn}$ and $^{176}\text{Hf}(n,2n)^{175}\text{Hf}$ ones. More details about these experimental measurements are presented in a separate contribution in this conference proceedings. Additional experimental measurements at energies above 15 MeV are planned for the near future, while statistical model calculations are in progress for all the above mentioned reactions.

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