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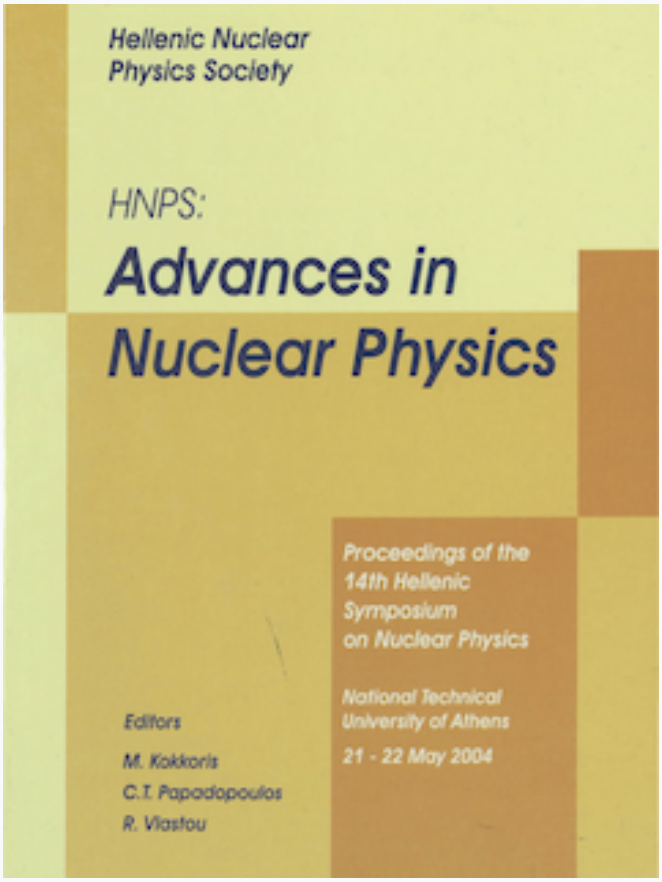

Measurement of the 241Am(n,2n) Reaction Cross Section, by the Activation Method

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Measurement of the $^{241}Am\,(n,2n)$ Reaction Cross Section, by the Activation Method

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Abstract

The measurement of the cross section of the reaction $^{241}Am(n,2n)^{240}Am$, has been performed, for the first time at neutron energies of 8.8, 9.6, 10.6 and 11.4MeV, by the activation method. The monoenergetic neutron beam was produced at the 5.5MV TANDEM accelerator of NCSR "Demokritos", using the $^{2}H(d,n)^{3}He$ reaction. During the 5-day long irradiation, the neutron flux was monitored by a BF_3 counter. The neutron yield as well as the beam current, were recorded in 100 sec intervals by two multiscaling units. The radioactive target consisted of a 37GBq ^{241}Am source enclosed in a Pb container. A natural Au foil, a ^{27}Al foil and a ^{93}Nb foil were used as reference materials for the neutron flux determination. The activity of the irradiated targets, was measured off-line by a 56 % relative efficiency, HPGe detector. The first preliminary results are presented and compared with evaluated libraries and existing ! data.

1 Introduction

Reactions of(n,xn) type are important for the development of fast reactors like the Accelerator Driven Systems (ADS) [1] , since the neutron balance in the core of the reactor, is affected by the neutron multiplication caused by such reactions. Americium is one of the most abundant isotopes in spent nuclear fuel, as well as the most highly radiotoxic among the actinides. Thus, the $^{241}Am\,(n,2n)$ reaction cross section is quite significant for the design of ADS systems. Furthermore, considering nuclear physics, the reaction $^{241}Am\,(n,2n)$ proceeds through the creation of the compound nuclei ^{242}Am, ^{241}Am, and

^{240}Am and allows study of nuclear models, in Americium isotopes lighter than ^{242}Am, which have not been investigated, since they can not be created by the better known reactions $^{241}Am\,(n,\gamma)$ and $^{241}Am\,(n,f)$. Even in the case of these reactions, there are large disc! repancies between measurements. These discrepancies, affect directly the predictions of evaluations for the reactions $(n,2n)$ and $(n,3n)$, since the better known reactions are used as constraints in the nuclear model calculations. Conversely, experimental data on the $(n,2n)$ or $(n,3n)$, could be used as constraint for resolving the discrepancy in the data for the other reactions. Until today, experimental data on the cross section of this reaction, span a limited energy range (13.9-15.1MeV [3,4]), while at the same time, theoretical predictions and evaluations for the cross section, differ in some energy regions by more than an order of magnitude. In this work, the cross section of the reaction $^{241}Am(n,2n)^{240}Am$ has been determined for the first time at the range of 8.8 to 11.4 MeV, by the activation method.

2 Experiment

2.1 Neutron Production

The measurements were performed at the TANDEM Accelerator of NCSR "Demokritos". The neutron beam was produced by the $^{2}H\,(d,n)^{3}\,He$ reaction, using a gas target which consisted of a stainless steel gas cell with a diameter of 1cm and a length of 3.7cm. A detailed description of the cell is given in [5]. A $5\mu m$ thick Mo foil served as the gas target window while a Pt foil served as the beam stop. At such a thickness, the molybdenum window allowed beam currents of up to $6\mu A$ to be used for many days of continuous irradiation, before the appearance of a pinhole leak. Furthermore, a gas pressure higher than 2000mbar could be used without breaking the window. The Pt foil, reduced the production of neutrons from the beam stop so that the resulting beam would be highly monoenergetic in most cases. During the irradiations, a cold air jet was used for cooling the cell. With this setup, a flux of the order of $2! - 4 \cdot 10^{6}\frac{n}{cm^{2}\cdot s}$ could be maintained for more than five days of continuous run.

2.2 Samples and preparation

The Americium target consisted of a 37GBq ($1\ Ci$) ^{241}Am source in the form of Americium oxide (AmO_2), encapsulated in stainless steel, provided by Isotope Products Blaseg GMBH. Apart from Americium, the target contained also, a quantity of ^{154}Eu, of the order of $100\mu Ci$ (370kBq). Since this was a

highly radioactive material, the Americium source was placed inside a lead cylindrical box with a wall thickness of 3 mm, properly shaped and sized so that the sample would fit exactly on its shielding. Thus, the greatest part of the gamma radiation produced by the target, coming in the form of a 59,6 keV gamma ray, was completely attenuated.
For the determination of the neutron beam flux, three thin foils made of reference materials —^{27}Al, ^{93}Nb and ^{197}Au—were placed in front of the Americium target. The diameter of the reference targets was exactly the same with that of the ^{241}Am source. Thus, the neutron flux at the position of the Americium sample, could be determined from the activity induced on the foils due to the reactions $^{27}Al(n,a)^{24}Na$, $^{197}Au(n,2n)^{196}Au$, and $^{93}Nb(n,2n)^{92m}Nb$.

2.3 Irradiation

The samples were placed for irradiation,at 0^{o}, at a distance of 9 cm from the center of the cell. In that position the angular acceptance of the targets was less than $\pm 8^{o}$ with respect to the beam. The energy spread of neutrons from the $^{2}H(d,n)$ reaction irradiating the target, was of the order of 50keV or less, for the netron energies used. The BF_3 monitor was placed at a distance of 2m away from the gas cell at an angle of 0°. During the irradiations, the pressure inside the gas cell was kept constant between 1000 and 1800 mbar. The deuteron beam current impinging on the gas target during the run, was kept between 3 and 6 μA. Each irradiation run lasted for about 100 h, in order to

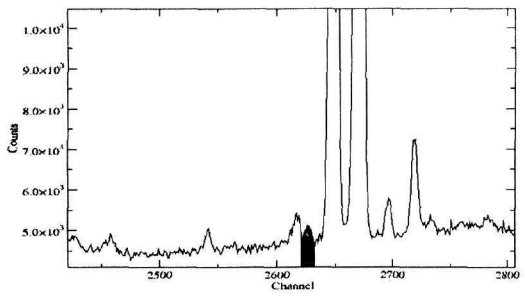

Fig. 1. *An enlarged view of the γ ray spectrum of the ^{241}Am target, at the area of interest for the activation measurement. The shaded peak corresponds to the 987.76 keV transition from the deexcitation of ^{240}Am, to ^{240}Pu. All the other peaks observed, are characteristic γ rays from ^{154}Eu contained in the target.*

accumulate on the Americium target, about 74% of the core activity. After the irradiation, the samples were transferred to the off-line gamma spectroscopy system for the activity measurement. A typical spectrum of the irradiated

Americium target, is presented in 1.
A BF_3 counter was used for monitoring the neutron beam. The output of the BF_3 detector was feeded into a multiscaling ADC system. The yield of the detector was registered every 100sec, producing at the end of the run a chart of the neutron beam fluctuations with respect to the irradiation time. A typical spectrum is presented in figure 2. At the same time, the integrated current of the deuteron beam at the position of the target, was registered for the same time interval, in a separate multiscaling ADC, producing the chart of fluctuations for the deuteron beam. This second chart was used for intercomparison purposes between the flux registered by the neutron monitor and the integrated current. The effect of the beam fluctuations to the evaluation of nuclei produced by the neutron irradiation is presented in Section 3.1.
After the irradiation, the activity of the samples was measured off-line by a system based on a HP Ge detector. The detector had an absolute relative efficiency of 56%. To reduce the contribution of background to the activity measurement, the detector was placed inside a specially prepared lead shield.

3 Analysis

The nucleus ^{240}Am produced by the reaction $^{241}Am\,(n,2n)^{240}\,Am$, decays by electron capture, to ^{240}Pu,with a half life of 50.8 h. It's dissintegration is accompanied by the emission of gamma rays. The most intense transitions from the deexcitation of ^{240}Pu, are at 888,8 keV, and 987,8 keV with intensities of 25.5% and 74% respectively [6]. Since the second one is much more intense, it was the most prominent for the activation measurement.
After the off-line measurements, the relevant peaks appearing in the spectra were analyzed, to determine the induced activity. All spectra were corrected for the dead time of the measurement, for solid angle effects and for the self absorption in the sample. The decay of the produced nuclei during the irradiation as well as for the time interval between the irradiation and the activity measurement, was also taken into account. The efficiency of the detector at the position of the activity measurements in the case of the reference targets, was determined using a calibrated ^{152}Eu source, at the same distance. A special procedure (described in detail in section 3.2) allowed for the simultaneous efficiency, solid angle and self absorption correction of the Americium target spectrum.

3.1 Determination of the cross section

The cross section σ, of the reaction $^{241}Am(n,2n)^{240}Am$, is given in terms of the number of ^{240}Am nuclei N_p produced during the irradiation interval, t_B,

in a target with a number of ^{241}Am nuclei, N_τ, by the equation

$$\sigma = \frac{N_p}{N_\tau} \cdot \frac{1}{\Phi} \tag{1}$$

provided the fluence of particles, $\Phi = \int_0^{t_B} f(t) \cdot dt$, is also known. The fluence of particles can be deduced by the activity induced to the reference foils. The equation for the cross section of the reaction then becomes,

$$\sigma = \frac{N_p}{N_\tau} \cdot \frac{N_t^r}{N_p^r} \cdot \sigma_r \tag{2}$$

where the quantities N_t^r and N_p^r are the target and produced nuclei for the reference foil, and σ_r, the cross section of the reference reaction. For the cross sections of the reference reactions, the values of the IRDF-2002 compilation by IAEA [2], were used.

3.2 Correction for the decay of the samples

The samples were corrected for the decay of nuclei between the end of irradiation and the activity measurement, using the analytical equations of radioactive decay. In order to correct the production of ^{240}Am nuclei in the samples for their decay during the irradiation, the competitition between creation and decay of nuclei had to be taken into account. This is given by the following differential equation:

$$\frac{dN}{dt} = \sigma \cdot \Phi(t) \cdot N_\tau - \lambda \cdot N \tag{3.3}$$

where for the case of the reaction $^{241}Am(n,2n)^{240}Am$:
N, is the number of ^{240}Am nuclei, and N_τ the number of ^{241}Am nuclei present in the target,
$\Phi(t)$, is the neutron beam flux at time t,
λ, is the decay constant of the ^{240}Am nucleus, and
σ, the cross section of the reaction (n,2n), that creates the nucleus.
The first term in equation 3.3 stands for the creation of nuclei by a particle flux Φ, while the second term, describes the decay of the produced nuclei with a decay constant λ. This differential equation has an analytic solution of the form:

$$N(t) = \frac{\int e^{\int \lambda \cdot dt} \cdot \sigma \cdot N_\tau \cdot \Phi(t) + C}{e^{\int \lambda \cdot dt}} \tag{3.4}$$

Using 3.4 to find the number of ^{240}Am nuclei at the end of the irradiation time τ_B, we end up with the relation:

$$N(T_B) = \sigma \cdot N_\tau \cdot \Phi_I \cdot \frac{\int_0^{T_B} e^{\lambda \cdot t} \Phi(t) \cdot dt}{\int_0^{T_B} \cdot \Phi(t) \cdot dt} \cdot e^{-\lambda T_B} \tag{3.5}$$

where $\Phi_I = \int_0^{T_B} \Phi(t) \cdot dt$ is the total fluence of the irradiation. The last term in 3.5

$$F_B = \frac{\int_0^{T_B} e^{\lambda \cdot t} \Phi(t) \cdot dt}{\int_0^{T_B} \cdot \Phi(t) \cdot dt} \cdot e^{-\lambda T_B} \tag{3.6}$$

is the correction factor for the decay of nuclei during the irradiation. For the ideal case of a stable neutron beam, it reduces to:

$$F_B = \frac{1 - e^{-\lambda T_B}}{\lambda T_B} \tag{3.7}$$

In the real situation however, of the fluctuating beam produced by the accelerator, 3.6 had to be solved numerically bin by bin, using the multiscaler time spectrum of the beam flux or/and the current inregrator . A typical spectrum from the multiscaler, is presented in figure 2.

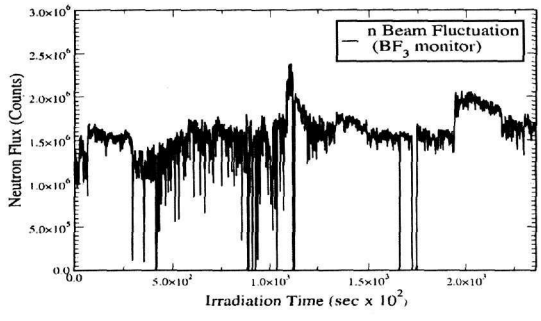

Fig. 2. *A typical chart of the neutron beam fluctuations*

3.3 Correction for solid angle and self absorption effects

The reference foil data, were corrected for the effect of the disk to disk geometry of the measurements, as well as for the self absorption of gamma rays in the target material. In the case of the Americium sample however, the

gamma rays produced by the target itself, were used to make the necessary corrections for the determination of the cross section. The ratio of produced to target nuclei is given by:

$$\frac{N_p}{N_t} \propto \frac{\frac{Y_p^i}{I_p^i} \cdot \frac{1}{\epsilon_\gamma^i f_{ss}^i}}{\frac{Y_t^j}{I_t^j} \cdot \frac{1}{\epsilon_\gamma^j f_{ss}^j}} \tag{3.8}$$

where:
Y_p^i is the yield of the photopeak with energy i of the produced nucleus,
Y_t^j is the yield of the photopeak with energy j of the target nucleus,
I_p^i, I_t^j are the corresponding intensities,
ϵ_γ are the corresponding efficiencies, and
f_{ss} the factor for self absorption and solid angle correction.
In the previous equation, for the special case where $i = j$, the efficiency and correction factors, cancel out, because the gamma rays of the produced and the target nucleus, are generated in the same sample —the Americium target—, subtend the same solid angle to the detector, and sustain the same absorption in the target. Thus, if the ratio Y_t/I_t is determined at the energy $i = j$, the ratio N_p/N_t can be determined without applying any corrections. The ratio Y_t/I_t, has been determined at the energies of 888,8 keV and 987,8 keV, by fitting it's dependence with the gamma ray energy, using peaks from the Americium target spectrum. A fit of the Y_t/I_t ratio for one of the measurements, is presented in figure 3.

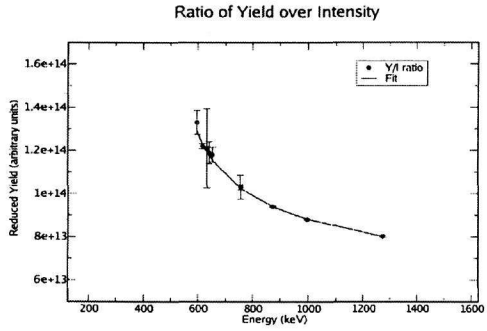

Fig. 3. Fit of the Y_t/I_t ratio using gamma rays of the target

4 Results

The cross section of the reaction $^{241}Am\,(n,2n)^{240}Am$, has been measured at the energies of 8.8, 9.6, 10.6 and 11.4 MeV, by the activation method. The results of the measurement are summarized in 4. The comparison of the experimental values obtained in this work with previous measurements [3,4] and predictions from the evaluated data libraries, clearly shows out that data on a broad energy range are really important for the resolution of discrepancies in nuclear data.

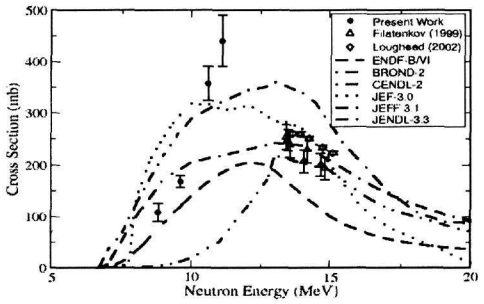

Fig. 4. The cross section of $^{241}Am\,(n,2n)$ reaction in comparison with recent evaluations and previous measurements [3,4]

Further measurements, both at higher and lower energies is planned to be carried out in the near future at NCSR "Demokritos". In addition, the reaction will be investigated from the theoretical point of view. For this purpose, statistical model calculations will be performed using the code STAPRE [7].

Acknowledgments

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