Measurement of the 241Am(n,2n)240Am cross section at 17.5 MeV

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Abstract

The $^{241}$Am$(n,2n)^{240}$Am reaction cross section has been measured at neutron beam energy 17.5 MeV, relative to the $^{27}$Al$(n,\alpha)^{24}$Na, $^{197}$Au$(n,2n)^{196}$Au and $^{93}$Nb$(n,2n)^{92}$m Nb reference reaction cross sections, using the activation technique. The irradiation was carried out at the Van der Graaff 5.5 MV Tandem accelerator laboratory of NCSR “Demokritos” with monoenergetic neutron beam provided by means of the $^3$H(d,n)$^4$He reaction, implementing a new Ti-tritiated target. The high purity Am target has been constructed at IRMM, Geel, Belgium and consisted of 40 mg $^{241}$Am in the form of AmO$_2$ pressed into pellet with Al$_2$O$_3$ and encapsulated into Al container. Due to this high radioactivity (5 GBq), the Am target was enclosed in a Pb container for safety reasons. After the end of the irradiation, the activity induced by the neutron beam at the target and reference foils, was measured off-line by two 100%, a 50% and a 16% relative efficiency, HPGe detectors.

Keywords: X-ray fluorescence, fundamental parameters method

1. Introduction

Studies of $(n,xn)$ reactions on minor actinides are of considerable significance, both for their importance to fundamental research in Nuclear Physics, as well as for practical applications, especially for the development of fast reactors since they affect the neutron balance in the reactor core. The study of $^{241}$Am$(n,2n)^{240}$Am reaction is important as Am is one of the most abundant isotopes in the spent fuel cycle and one of the most highly radiotoxic among the actinides. Five recent works provide data from threshold to 15 MeV [1, 6], but one of these works provided data above 16 MeV [5]. In order to enrich this energy region with more data, a new measurement has been performed at the 5.5 MV Tandem T11/25 accelerator laboratory of NCSR “Demokritos”, by using the activation method.

2. Experimental Process

In the present work, the cross section measurement of $^{241}$Am$(n,2n)^{240}$Am threshold reaction has been performed at neutron beam energy 17.5 MeV at the 5.5 MV Tandem T11/25 accelerator laboratory of NCSR “Demokritos”, by using the activation method. The neutron beam has been produced by means of the $^3$H(d,n)$^4$He reaction implementing a new Ti-tritiated target consisted of 2.1 mg/cm$^2$ Ti-t layer on a 1 mm thick Cu backing for good heat conduction. The activity of the tritium target is 373 GBq. The deuterons were accelerated to 2.5 MeV and passed though two 5 $\mu$m Mo foils in order to degrade their energy to 1.5 MeV, where the cross section of the $^3$H(d,n)$^4$He reaction is high enough to produce neutron beam at a flux of the order of $6 \cdot 10^7$ n/(sec $\cdot$ cm$^2$). The deuteron beam current was $\approx 1$ $\mu$A and the neutron flux was monitored by a BF$_3$ detector placed at a distance of 3 m from the neutron source. The accumulated spectra were stored at regular time intervals to account for the beam fluctuations in the off-line analysis of the data. The flange with the tritium target assembly was air cooled during the deuteron irradiation.
The Am target has been provided by IRMM, Belgium, coming from the same batch of targets used by Sage et al. [5]. It consists of 40 mg $^{241}$Am in the form of AmO$_2$ pressed into pellet with Al$_2$O$_3$ and encapsulated into Al container. Due to its high radioactivity (5 GBq), the Am sample was placed inside a 3mm lead cylindrical shielding and high purity foils of Al were placed at the front and the back of the shielding, along with the Au and Nb foils at the back, to monitor the neutron flux and to account for its variation with distance. The target assembly was placed at about 2 cm from the tritium target, as shown in Fig. 1, thus limiting the angular acceptance to $\pm 15^\circ$, where the produced neutrons are practically isotropic and monoenergetic.

The activation lasted for 4 days, corresponding to 2 half lives of the residual nucleus $^{240}$Am. The induced activity of product radionuclides in Am target and reference foils was measured with four HPGe detectors of 100%, 100%, 56% and 16% relative efficiency, properly shielded with lead blocks in order to reduce the contribution of the natural radioactivity. Two Am gamma-ray spectra were taken at a distance of approximately 7 cm from the 100% relative efficiency HPGe detector, before and after the irradiation (as shown in Fig. 2), to ensure that there is no contamination in the 987.8 keV photopeak from the decay of the $^{240}$Am residual nucleus. The two spectra clearly show that the 987.8 keV $\gamma$-ray is free from interference by the sample activity. The second characteristic transition from the deexcitation of $^{240}$Am at 888.9 keV is contaminated, thus only the 987.8 keV $\gamma$-ray was used for cross section analysis.

3. Data Analysis and Results

The efficiency of the detection setup, including the extended geometry of the Am sample and self-absorption effects, was extracted by using an experimental technique, which is based on the activity of the Am target as described in Ref. [3]. The ratio $R = N_p/N_t$ can be determined experimentally from the gamma rays emitted by the target $^{241}$Am nuclei $N_t$, and produced $^{240}$Am nuclei $N_p$, as a function of the gamma-ray energy, as shown in Fig. 3.

From this plot, the ratio $R$ for the 987.8 keV can be extracted and subsequently used for the determination of the cross section of the $^{241}$Am(n,2n)$^{240}$Am reaction according to the following expressions:

$$\sigma = \frac{N_p}{N_t} \cdot \frac{1}{\Phi_{Am}} \Rightarrow \sigma = R \cdot \frac{1}{\Phi_{Am}}$$

As for the neutron beam flux in the Am pellet, MCNP5 [7] Monte Carlo simulations were carried out in order to reproduce the experimental values of the flux deduced from the reference foils Al, Au and Nb shown.
Figure 2: High-energy part of the $\gamma$-ray spectra for non-irradiated and irradiated Am sample at 17.5 MeV

Figure 3: High-energy part of the $\gamma$-ray spectra for non-irradiated and irradiated Am sample at 17.5 MeV
Table 1: Experimental neutron flux at 17.5 MeV and the simulated flux in the middle of the Am pellet

<table>
<thead>
<tr>
<th>Target</th>
<th>$\Phi \times 10^{11}$ n/cm$^2$</th>
<th>$\delta\Phi \times 10^{11}$ n/cm$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al front</td>
<td>2.98</td>
<td>0.13</td>
</tr>
<tr>
<td>Am pellet</td>
<td>1.98</td>
<td>0.10</td>
</tr>
<tr>
<td>Al back</td>
<td>1.05</td>
<td>0.05</td>
</tr>
<tr>
<td>Au</td>
<td>0.88</td>
<td>0.04</td>
</tr>
<tr>
<td>Nb</td>
<td>0.84</td>
<td>0.04</td>
</tr>
<tr>
<td>Al</td>
<td>0.80</td>
<td>0.04</td>
</tr>
</tbody>
</table>

Figure 4: Cross section of the $^{241}$Am(n,2n) reaction

in Table 1. The experimental flux at the Al foil in front of the Am target (Al front) was used as starting flux, while the geometry of the whole Am target assembly was described in detail for the simulations. The simulated neutron flux reproduced fairly well the experimental one at the back foils (Al back, Au, Nb, Al), certifying the validity of the simulations. Thus, the simulated flux in the middle of the Am pellet, shown also in Table 1, was used to deduce the cross section.

The $^{241}$Am(n,2n)$^{240}$Am cross section at 17.5 MeV, extracted by this method, is plotted in Fig. 4 along with other data from literature and seems to agree with the data by Sage et al. [5] within its experimental error.

4. Summary

The $^{241}$Am(n,2n)$^{240}$Am reaction cross section has been measured at neutron beam energy 17.5 MeV, relative to the $^{27}$Al(n,α)$^{24}$Na, $^{197}$Au(n,2n)$^{196}$Au and $^{93}$Nb(n,2n)$^{92}$mNb reference reactions, implementing the activation technique. The purpose of this measurement was to provide more experimental data in the energy region above 16 MeV, where only one [5] of the five recent works (shown in Fig. 4) presented data until now. The irradiation was carried out at the Van der Graaff 5.5 MV Tandem T11/25 accelerator laboratory of NCSR “Demokritos”, by using the quasi-monoenergetic neutron beam produced via the $^3$H(d,n)$^4$He reaction.
at a flux of order $6 \cdot 10^5 \, n/(cm^2 \cdot sec)$. The Am target consisted of 40 mg $^{241}$Am in the form of AmO$_2$ of high purity. The induced gamma-ray activity of the Am target and the reference foils was measured off-line with high resolution HPGe detectors. The deduced cross section value agrees with the data by Sage et al. [5] within its experimental error.

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