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New measurement of the $^{241}\text{Am}(n,2n)^{240}\text{Am}$ cross section

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Abstract

Cross section for $^{241}\text{Am}(n,2n)^{240}\text{Am}$ reaction has been measured at the VdG Tandem accelerator of NCSR "Demokritos", at neutron beam energy 10.4 MeV, using the activation technique. The high purity and high radioactivity (5GBq) Am target has been constructed at IRMM, Belgium and consisted of 40 mg Am in the form of AmO₂ pressed into pellet with Al₂O₃ and encapsulated into Al container. The absolute flux of the beam was obtained with respect to the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reference reaction. The induced gamma-ray activity of ^{240}Am and ^{24}Na was measured with high resolution HPGe detectors.

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 $^{241}\text{Am}(n,2n)^{240}\text{Am}$ 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 20 MeV, with severe discrepancies among them in the energy region 10 to 12 MeV. [1–5]. Around 14 MeV, the data of Loughheed et al. [2] and Tonchev et al. [4] are in good agreement, while the data by Filatenkov et al. [1] are systematically lower by about two standard deviations. The data by Perdikakis et al. [3], have been measured at NCSR "Demokritos" from 8.8 to 11.4 MeV, and below 10 MeV agree well with both sets of data by Tonchev et al. [4] and Sage et al. [5], while in the energy region 10 to 12 MeV are

much higher than the measurements of [3] and [4]. In order to resolve these discrepancies between the existing data, new measurements have been performed at the 5 MV Tandem T11/25 accelerator laboratory of NCSR "Demokritos" with the activation method implementing a high purity Am target provided by IRMM.

2. Experimental Process and Results

Cross section measurements of (n,2n) threshold reaction on ^{241}Am has been performed at neutron beam energy 10.4 MeV at the 5 MV Tandem T11/25 accelerator laboratory of NCSR "Demokritos", by using the activation method. Quasi-monoenergetic neutron beams were produced via the $^2\text{H}(\text{d},\text{n})^3\text{He}$ reaction at a flux of the order of $\sim 10^6 \text{ n}/(\text{cm}^2 \text{ sec})$. The Am target has been provided by IRMM, Belgium, coming from the same batch of targets used by Sage et al. [5]. It consisted of 40 mg Am in the form of AmO_2 pressed into pellet with Al_2O_3 and encapsulated into Al container. Due to its high radioactivity ($\sim 5\text{GBq}$), the Am target was placed inside a 3mm lead cylindrical shielding and was irradiated with 10.4 MeV neutrons for three days. The absolute flux of the beam was obtained with respect to the $^{27}\text{Al}(\text{n},\alpha)^{24}\text{Na}$ reference reaction, while its variation was monitored by a BF_3 detector placed at a distance of 3 m from the neutron source. In addition, the $^{93}\text{Nb}(\text{n},2\text{n})^{92\text{m}}\text{Nb}$ and $^{197}\text{Au}(\text{n},2\text{n})^{196}\text{Au}$ reference reactions were used in order to estimate more accurately the neutron flux in the Am pellet, implementing Monte Carlo simulations. Thus, high purity foils of Al were placed at the front and the back of the Am sample lead shielding, along with Au and Nb foils at the back.

The induced activity of product radionuclides of both Am target and reference foils was measured with HPGe detectors of 80% and 56% efficiency. Gamma-ray spectra were taken at a distance of $\sim 10\text{cm}$ from the Ge detectors, while for Am target spectra were taken both before and after the irradiation, to ensure that there is no contamination in the 987.8keV photopeak corresponding to the characteristic gamma ray from the decay of the ^{240}Am residual nucleus. The efficiency of the detection setup, including the extended geometry of the Am sample and self absorption effects, was extracted by using two different techniques, an experimental method taking into account the natural activity of the ^{241}Am sample as described in [3] and a simulated one implementing the MCNP [6] code. The simulated efficiency was deduced via a series of MCNP calculations which helped to fix the various geometrical parameters involved, by reproducing the experimental spectra taken for different reference setups, implementing a calibrated ^{152}Eu point source placed at 10cm distance from the HPGe detector: (a) Ge detector and source to fix the detector geometry, its dead layer, Al window etc. (b) Ge detector and source with the lead cylindrical box in front to fix the shielding, (c) Ge detector and source at the back of the

lead cylindrical box with the Al container inside, to fix the container and (d) Ge detector and Am sample in its shielding before irradiation to reproduce the expected density of Am. All these experimental spectra were reproduced fairly well by the MCNP simulations and the deduced parameters were then used to simulate the efficiency for the 987.8 keV gamma ray. The preliminary results for the cross section determination are (345 ± 3) mb from the experimental method and (232 ± 24) mb from the method using the MCNP simulations to estimate the efficiency of the detection system.

The mean cross section value from the two methods at 10.4 MeV is plotted in Fig. 1 along with other data from literature and seems to agree with the data by Tonchev et al. [4] within their experimental errors. As for the old data by Perdikakis et al. [3], which agreed very well with the Tonchev data [4] at 8.8 and 9.6 MeV while above 10 MeV the cross section values were much higher, this discrepancy could be attributed to the contaminations of the Am target, mainly by ^{154}Eu , used in ref. [3]. It is possible that above 10 MeV, a neutron induced reaction on the contaminants of the target is open and produces a gamma ray which interferes with the 987.8 keV transition, thus amplifying its yield and increasing the cross section value.

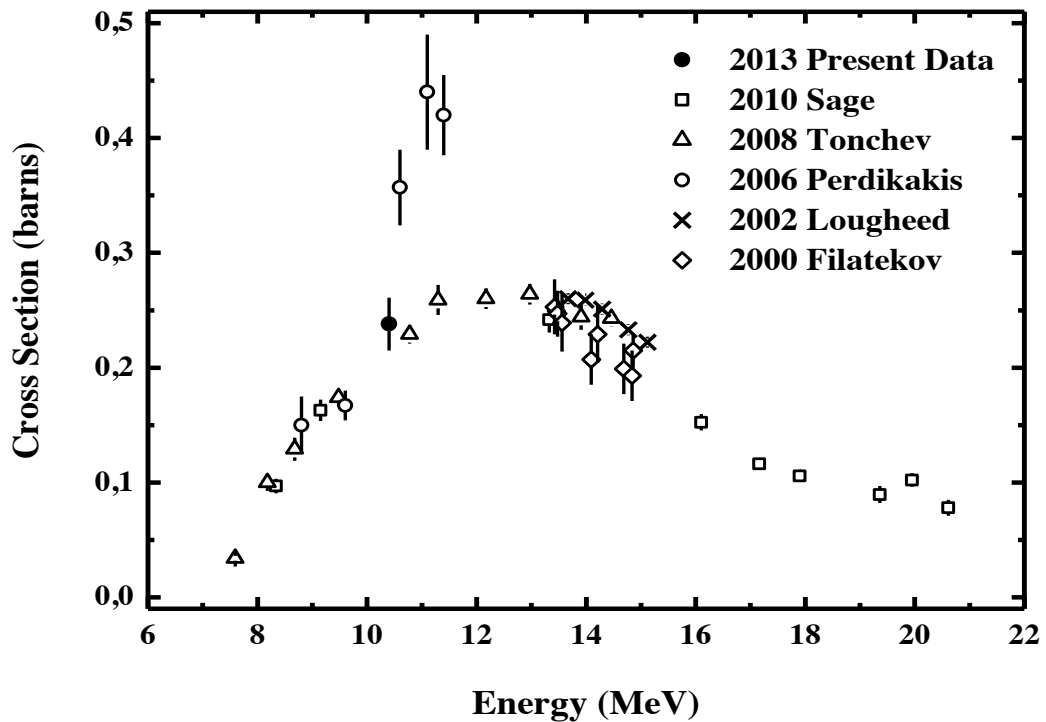


Fig. 1. Cross section of the $^{241}\text{Am}(n,2n)$ reaction

3. Summary

The cross section of the $^{241}\text{Am}(n,2n)^{240}\text{Am}$ reaction at 10.4 MeV has been measured with respect to the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$, $^{93}\text{Nb}(n,2n)^{92\text{m}}\text{Nb}$ and $^{197}\text{Au}(n,2n)^{196}\text{Au}$ reference reactions, implementing the activation technique. The purpose of these measurements was to resolve the discrepancies between the data by Perdikakis et al. [3] and by Tonchev et al. [4] which agree below 10 MeV, while above 10 MeV they differ by a factor of about 2. The irradiation was carried out at the 5 MV Tandem T11/25 accelerator laboratory of NCSR "Demokritos", by using the quasi-monoenergetic neutron beam produced via the $^2\text{H}(d,n)^3\text{He}$ reaction at a flux of the order of $\sim 10^6$ n/(cm²sec). The Am target consisted of 40 mg Am in the form of AmO₂ of high purity. The induced gamma-ray activity of the Am target and the reference foils was measured with high resolution HPGe detectors. The deduced cross section value agrees with the data by Tonchev et al. [4] within their experimental errors, meaning that the high cross section values of previous data by Perdikakis et al. [3] measured in the same facility, are due to the contaminations of the Am target. These contaminants could induce threshold reactions above 10 MeV producing a gamma ray which interferes with the 987.8 keV transition. More measurements are planned to be carried out in the near future at higher neutron beam energies.

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