Measurement of the ^{241}Am$(n, 2n)$ reaction cross section using the activation method

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In the context of the n_TOF Collaboration, 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 from 8.8 to 11.4 MeV, by the activation method, relative to the ^{27}Al$(n, a)^{24}$Na reaction reference cross section. The monoenergetic neutron beam was produced at the 5.5 MV TANDEM accelerator of NCSR "Demokritos," by means of the ^{2}H$(d, n)^{3}$He reaction, using a deuterium filled gas cell. The radioactive target consisted of a 37 GBq ^{241}Am source enclosed in a Pb container. After the end of the irradiation, the activity induced by the neutron beam at the target and reference, was measured off-line by a 56% relative efficiency, HPGe detector.

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Americium is an important isotope for transmutation and the development of "clean" nuclear power systems. It is one of the most abundant isotopes in spent nuclear fuel, as well as one of the most highly radiotoxic among the actinides [1]. Furthermore, reactions of (n, xn) type are important for the development of fast reactors like the Accelerator Driven Systems (ADS) [2–6], since the neutron balance in the core of the reactor, is affected by the neutron multiplication caused by such reactions. The alteration of the neutron spectrum caused by americium (and other isotopes contained in spent nuclear fuel) in the core of such a reactor, the transmutation of nuclear waste through fission and the efficiency of energy production, could be affected by the cross sections of (n, xn) and (n, xnf) reactions. However, there is quite a lack of experimental data concerning these reactions for ^{241}Am, and theoretical calculations are used instead. Furthermore, theoretical evaluations of the ^{241}Am$(n, 2n)$ reaction cross section, present significant discrepancies (Fig. 1), implying that the experimental determination of the cross section, is important for the design of ADS systems as well as for theoretical statistical model calculations. The reliability of such calculations, is affected by uncertainties in the theoretical description of the contribution of multiple chance fission channels ^{241}Am(n, xnf), to the total fission cross section ^{241}Am(n, F). It has to be noted, that the consistent theoretical description of (n, xn) and fission reactions of ^{238}U and ^{232}Th nuclei, has been successfully utilized in Refs. [7–9], to reduce the uncertainties in the corresponding statistical model calculations. Until the present, experimental data on the cross section of the ^{241}Am$(n, 2n)$ reaction spanned a limited energy range (13.9–15.1 MeV [10,11]).

In the present work, the cross section of the reaction ^{241}Am$(n, 2n)^{240}$Am has been measured for the first time in the energy range of 8.8 to 11.4 MeV, by the activation method. The cross section has been determined by means of the 987.8 keV gamma ray, from the deexcitation of ^{240}Am. The americium target consisted of a 37 GBq (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 100μCi (370 kBq). Special care was taken for the safe handling of the target. 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. The neutron fluence for the cross section measurement was determined by using the monitor reaction ^{27}Al$(n, \alpha)^{24}$Na as a reference. The induced γ activities, were determined by an absolute off-line measurement using a 56% HPGe detector.

The irradiations were performed at the 5.5 MV TANDEM Accelerator of NCSR "Demokritos." The neutron beam was produced by the reaction of the deuteron beam provided by the accelerator, with a deuterium gas target, utilizing the reaction ^{2}H$(d, n)^{3}$He. The gas target consisted of a stainless steel gas cell with a diameter of 1 cm and a length of 3.7 cm. A detailed description of the cell is given in Ref. [12]. A 5μm thick molybdenum foil served as the gas target window while a platinum foil served as the beam stop. During the irradiations, a cold air jet was used for cooling the cell at the position of the Mo window and the Pt beam stop. In this way, the heat transferred on the gas target by the deuteron beam, was dissipated effectively, in order to minimize the effect of heating to the deuterium gas pressure. With this setup, a flux of the order of $2 - 4 \times 10^{6}\, n/\mathrm{cm}^2\,\mathrm{s}$, could be maintained for more than 5 d of continuous run.

During the irradiations, a BF_3 counter placed at a distance of 2 m away from the gas cell at an angle of 0°, was used for monitoring the neutron beam fluctuations. The output of the BF_3 detector was fed into a multiscaling ADC system. The yield of the detector was registered every 100 s, producing at the end of the run a chart of the neutron beam fluctuations with respect to the irradiation time. The samples were placed for irradiation, at 0°, at a distance of 9 cm from the center of the cell. In that position the angular acceptance of the targets was

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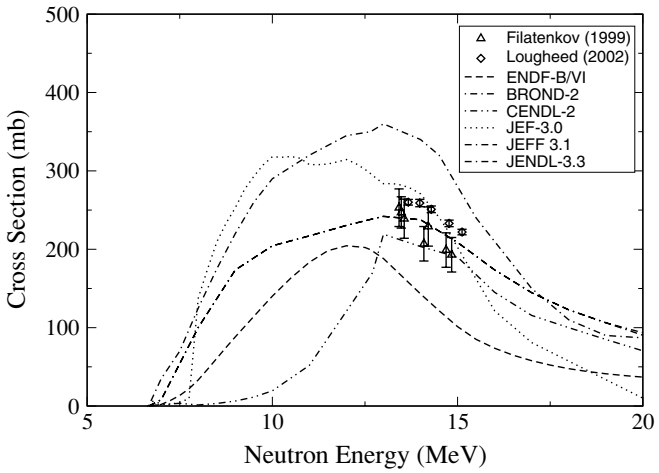

FIG. 1. Previous experimental data compared to evaluations from the major data libraries.

less than $\pm 8^{\circ}$ with respect to the beam. During the irradiations, the gas cell was operated at pressures ranging from 1000 to 1800 mbar depending on the energy of the deuteron beam, in order to keep the uncertainty in the neutron energy as low as possible. 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 accumulate approximately 74% of the core activity on the americium target. After the irradiation, the samples were transferred to the off-line gamma spectroscopy system for the activity measurement. The activity of the targets, was measured at distances of more than 15 cm from the detector face, so that coincidence summing effects could be considered negligible. For each measurement, the absolute efficiency was measured using a calibrated ^{152}Eu source, at the counting position.

The neutron energy distribution at the target position during the irradiations, was determined by taking into account the energy loss and straggling of the deuteron beam at the cell entrance foil and at the deuterium gas, using the analytical approach described by Klein *et al.* [13]. The results of this analytical procedure for the beam energy loss have been cross-checked against the results of the SRIM-2003 Monte Carlo code [14] for the energy loss of the deuteron beam in Mo (entrance window) and in the deuterium gas. The low production of parasitic neutrons from the gas cell window and beam stop and the monochromaticity of the neutron beam at the energies of measurement, has been verified experimentally by sequential gas-in and gas-out activation measurements, using natural Fe and ^{27}Al foils. The reduction of the neutron flux caused by the lead shielding of the americium target, was measured experimentally with activated aluminum foils at the outer and inner surfaces of the lead box, and within 5% the flux reduction has been found to be negligible. This experimental result is in agreement with the Monte Carlo calculation using the code MCNP-4c [15], that has been employed, in order to determine the energy and flux distribution of neutrons on each sample, as well as the contribution of scattered neutrons at energies above the reaction threshold, to the total neutron flux on the americium target.

Since the cross section σ of a neutron induced reaction at an incident energy E_n, is given in terms of the number of nuclei produced during the irradiation N_p, and the number of target nuclei N_τ, by the equation

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

in which the integrated neutron flux (fluence) Φ for the irradiation time t_B, is given in terms of the instant flux $f(t)$, by the integral

$$\Phi = \int_0^{t_B} f(t)dt. \tag{2}$$

The quantity N_p, has been deduced from the number of gamma ray events N_γ, registered in the Ge detector, by the expression

$$N_p(t_B) = \frac{N_\gamma}{\epsilon F I_\gamma D f_c} \tag{3}$$

taking into account for the decay of the produced nuclide, the fluctuation of the neutron beam flux over the irradiation interval t_B, through the factor

$$f_c = \frac{e^{-\lambda t_B} \int_0^{t_B} e^{\lambda t} f(t)dt}{\int_0^{t_B} f(t)dt}. \tag{4}$$

In Eq. (3), $D = e^{-\lambda t_1} - e^{-\lambda t_2}$ is the counting collection factor, with t_1, t_2, being the time intervals from the end of the irradiation, to the beginning and the end of the gamma ray counting with the Ge detector, respectively. In the same equation, I_γ is the intensity of the counted characteristic gamma ray, ϵ is the efficiency of the Ge detector, and F is the total correction factor to account for self absorption in the sample, and counting geometry.

It has to be noted, that in the case of the americium sample, an experimental method was preferred to the theoretical evaluation of the factor F, due to the rather complicated target and shielding layout. More specifically, the factor F was evaluated indirectly, through the determination of the ratio N_p/N_τ of Eq. (1) for the reaction ^{241}Am$(n, 2n)^{240}$Am, from the gamma rays emitted by the irradiated americium sample. While the number of produced nuclei is given by Eq. (3), the number of target (^{241}Am) nuclei, is given by an equivalent relation, without the f_c factor:

$$N_\tau = \frac{N'_\gamma}{\epsilon' F' I'_\gamma D'}. \tag{5}$$

Thus, the ratio N_p/N_τ, is given by the expression

$$\frac{N_p}{N_\tau} = \frac{N_\gamma}{I_\gamma} \cdot \frac{I'_\gamma}{N'_\gamma} \cdot \frac{D'}{D} \cdot \frac{1}{f_c} \cdot \frac{\epsilon' F'}{\epsilon F} = R \cdot \frac{\epsilon' F'}{\epsilon F}. \tag{6}$$

The quantity R can be determined experimentally from the gamma rays emitted by the target and produced nuclei. Since both are contained in the same sample, the gamma rays used for the determination of the number of ^{240}Am and ^{241}Am nuclei, are subject to absorption by an identical matrix of materials in the americium target, in an identical source to detector geometry. For a fixed energy, the overall correction factors F and F', are therefore, identical. The same holds for the detection efficiency, thus simplifying $N_p/N_\tau = R$.

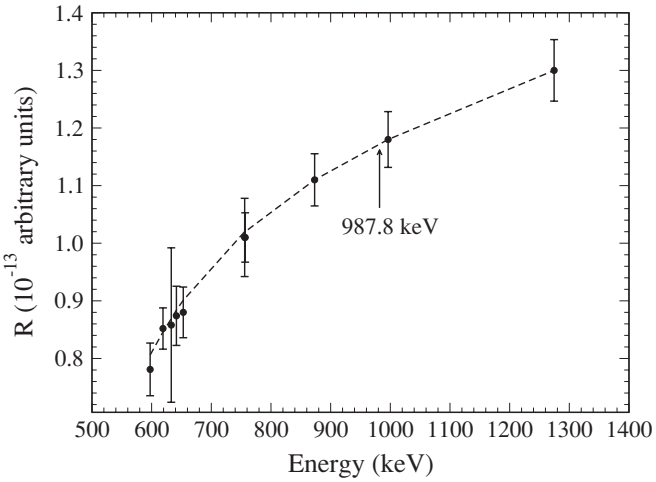

FIG. 2. Plot of the quantity R of Eq. (6), as a function of energy, taken at 11.4 MeV. The arrow denotes the point at 987.8 keV used for the reaction rate evaluation.

For the experimental determination of N_p/N_τ, the R ratio has been evaluated by measuring N'_γ of several gamma ray lines emitted above 500 keV from the ^{241}Am of the sample, as well as N_γ of the 987.8 keV characteristic gamma ray from the deexcitation of ^{240}Am. The presence of ^{154}Eu on the target has also been implemented, to cover the higher energy gamma ray region. The intensities I_γ of the gamma rays, were taken from Refs. [16–18]. The results are shown in Fig. 2, from which the value of R is taken for the 987.8 keV energy and subsequently used for the cross section calculation [Eq. (1)]. The activity of the ^{241}Am target has also been used in Ref. [10], for the determination of the number of target nuclei, producing very accurate results compared to other techniques.

For the 987.8 keV characteristic gamma ray of ^{240}Am, the evolution of the photopeak net area with time, has been plotted in Fig. 3, for each irradiation energy. The fair agreement of the experimental data with the fit corresponding to the

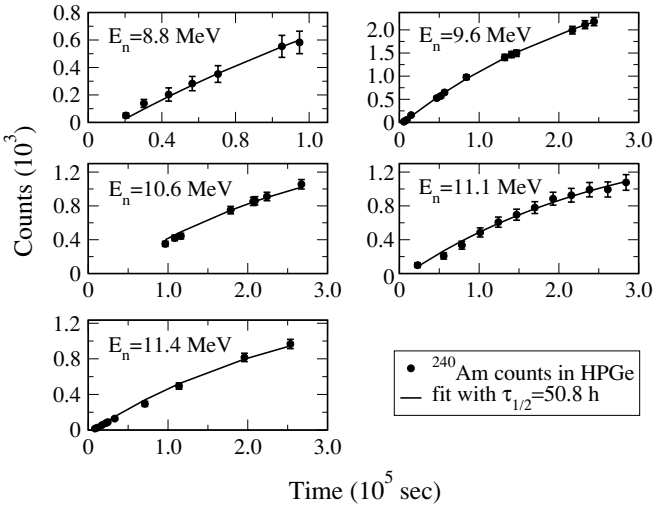

FIG. 3. Plot of the accumulated yield for the 987.8 keV characteristic gamma ray of ^{240}Am.

TABLE I. Cross section values for the reaction ^{241}Am$(n, 2n)^{240}$Am, deduced from this work.

Energy (MeV)	Cross section (mb)	Total error (mb)
8.8	150	25
9.6	167	13
10.6	357	33
11.1	440	50
11.4	420	35

50.8 h half-life of ^{240}Am [18], suggests that no contamination was present in the photopeak area. The transition from the deexcitation of ^{240}Am at $E_\gamma = 888.8$ keV [18], could not be used for the deduction of accurate results, since, apart from its lower intensity, it is adjacent to the 887.3 keV gamma ray from the deexcitation of ^{241}Am. It has to be noted that, although the intensity of most of the characteristic gamma rays emitted by the radioactive target is of the order of 10^{-6}%–10^{-7}%, due to the high activity of the sample, the photopeaks are prominent in the spectrum. However, at the maximum cross section energy of 11.4 MeV, it has been possible to analyse the 888.8 keV transition in the spectrum and determine experimentally the value of 0.4 ± 0.1 for the ratio of intensities between the 888.8 keV and 987.8 keV gamma rays. This value, is in agreement within the rather large experimental errors, to the value of 0.34 ± 0.03, deduced from the tabulated intensities.

Since the measurements were carried out with respect to a reference reaction, the quantity Φ in Eq. (1), can be substituted by

$$\Phi = \frac{N_p^r}{N_\tau^r} \cdot \frac{1}{\sigma^r} \tag{7}$$

depending on the number of target and produced nuclei on the reference foils N_τ^r and N_p^r, respectively. The cross section is

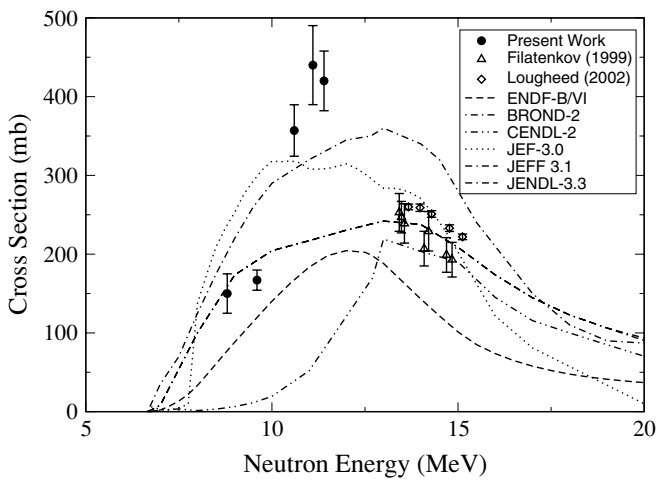

FIG. 4. Presentation of our data in comparison with previous measurements and evaluations by the major data libraries.

then given by the expression

$$\sigma = \frac{N_p}{N_\tau} \cdot \frac{N_\tau^r}{N_p^r} \cdot \sigma^r. \quad (8)$$

For the cross section of the reference reaction, the values of the IRDF-2002 compilation by IAEA [19], were used.

The results for the cross section are presented in Table I. Previous measurements and evaluations are presented in Fig. 4, in comparison with the results of this work.

The errors in the cross section table come from standard cross section values (1–2%), counting statistics (1–14%), decay data (0.2–3%), Monte Carlo and analytical simulation of irradiation conditions (5%), samples weight (1%), neutron fluence (3%), neutron energy determination (1–2%) and detector efficiency (5%).

The trend of the data is characterized by a steep increase of the experimental cross section from 8.8 to 11 MeV. Similar rapid rise has also been observed in the experimental cross sections of the reactions ^{238}U$(n, 2n)$ [20,21], ^{232}Th$(n, 2n)$ [22, 23], and ^{237}Np$(n, 2n)$ [24]. The energy at which the maximum value of the cross section appears cannot be determined with certainty from the experimental data presented in this work. The same holds for the plateau in the cross section, predicted by some evaluations (Fig. 4). For these questions to be answered, further measurements at the energy region between 11 and 13 MeV would be required.

In summary, the cross section of the reaction ^{241}Am$(n, 2n)^{240}$Am has been measured in the energy range from 8.8 to 11.4 MeV by the activation method. The data are particularly important for calculations within the framework of the Hauser-Feshbach model, especially in connection to the discrepancies in the ^{241}Am(n, F) cross section. Such calculations are currently under way using the code STAPRE [25].

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