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Progress Report on Research Contract HUN14318 for the CRP on
Minor Actinide Neutron Reaction Data (MANREAD)

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Abstract

The ground state cross section of ^{242}Am has been measured with beams of cold neutrons at the Budapest Research Reactor using the X-ray emission of the decay product of ^{242}Pu. This methodology avoids the uncertainty caused by resonance neutrons in the pile activations. The target was characterized with gamma and X-ray spectrometry. The obtained ground state cross section is 540 ± 32 b, which is at the low end of the most recent literature values, but agrees with most of them within their uncertainty.

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ABSTRACT

The ground state radiative neutron capture cross section of ^{241}Am has been measured with beams of cold neutrons at the Budapest Research Reactor using the X-ray emission of the decay product of ^{242}Pu. This methodology avoids the uncertainty caused by resonance neutrons in the pile activations. The target was characterized with gamma and X-ray spectrometry. The obtained ground state cross section is 540 ± 32 b, which is at the low end of the most recent literature values, but agrees with most of them within their uncertainty.

1. INTRODUCTION

Increasing amount of highly radioactive nuclear waste is one of the major concerns in nuclear industry. To decrease their amounts, transmutation and closed fuel cycles are considered in ADS and GEN-IV systems. Design of these systems requires better accuracy of nuclear data, especially for minor actinides. A long-standing problem is the improvement of the cross section uncertainties of the ^{241}Am, which is one of the most radiotoxic isotopes among the minor actinides.

Bouland published an excellent review on the contradictions between the available data [1] and initiated a series of new measurements to improve the cross section data for ^{241}Am.

At the cold beam facility of the Budapest Research Reactor (BRR) we can activate isotopes with a sub-thermal neutron beam that helps to avoid the complications arising from the lowest energy resonance (0.3 eV) for thermal or mixed neutron-field activations. Wisshak et al. has already used sub-thermal beam of neutrons for activation, and measured the activity of the ^{242}Am using electron spectrometry [2].

Target preparation for the various concurrently running experiments poses a difficult problem due to the lack of experienced radio-chemists.

Here we report our experimental results on the ground state radiative neutron capture cross section of ^{241}Am performed on a special metallic target obtained from the inventory of University of Debrecen, Hungary.

1.1 Present status of the thermal cross section data for ^{241}Am

In the ^{241}Am capture process both ^{242g}Am and ^{242m}Am are fed. The experimental and evaluated cross sections for the ground state cross section are presented in Table 1. As it can be seen from the table, the values are spreading between 532-768 b not considering the outlier value reported by Street et al. TOF values seems to scatter around 600 b, except the value from Weston et al. [3]. The data from pile activations vary much more presumably due to the difficulties in separation of the thermal and resonance neutron yields.

To avoid this Wissahak et al. used neutrons with a well defined energy [2] to measure the thermal cross section assuming 1/v behavior of the cross section of ^{241}Am for low energy. Their value seems to support lower ground state cross sections (see Table 1).

To clarify the situation we decided to perform measurements at our cold neutron beam facility with the method suggested by Bouland [1], i.e. the detection of the ^{242}Pu X-rays at 99.5 keV and 103.4 keV. This provides an independent detection technique of the decaying ^{242}Am ground state from the earlier experiments.

Table 1. Experimental and compiled thermal capture cross sections for ^{241g}Am. When only total cross sections were measured the ground state cross sections were calculated using the branching ratio of 0.914(7) of Fioni et al. [4].

Experiment	σ_{th} g.s. (b)	Unc.(b)	Method
Jandel et al. 2008 [5]	608	30	TOF calc. from total
Nakamura et al. 2007 [6]	628	22	Pile activation
Fioni et al. 2001 [4]	636	46	Pile activation
Maidana et al. 2001 [7]	602	9	Pile activation
Shinohara et al. 1997 [8]	768	58	Pile activation
Wisshak et al. 1982 [2]	571	33	Beam activation at 14.75 meV
Kalebin et al. 1976 [9]	620	20	TOF
Weston et al. 1976 [3]	532	46	TOF
Harbour et al. 1973 [10]	748	20	Pile activation
Dovbenko et al. 1971 [11]	598	95	TOF
Hoff et al. 1959 [12]	620	65	Pile activation
Pomerance 1955 [13]	571	32	Pile oscillation
Street et al. 1952 [14]	274	0	Pile activation
Hanna et al. 1951 [15]	568	57	Pile activation
Mughabghab 2006 [16]	535	11	
JEFF-3.1b 2006	591	29	
ENDF/B-VII.0c 2006	567	12	
JENDL-3.3 2002	584	0	

2. DESCRIPTION FACILITY

The activation of the target was made at the in-beam Mössbauer station of the Budapest Research Reactor. This station has been equipped with a tapered focusing guide. The thermal equivalent neutron flux was measured to be about 10^9 n cm^2 s^{-1}. The entrance window of this guide section is 1.5×1.5 cm^2. The sample was directly taped to the end window of the guide.

The activated samples were measured with a 200 mm^2 active area LeGe detector (CANBERRA GL0215R). The detector has a thin (0.15 mm) Be-window and a resolution of 0.5 keV at 100 keV. The detector and the sample were placed in a lead cage to decrease the ambient background. The detector signal was analyzed with a PIXIE-4 type digital analyzer and the data was collected on a dedicated PC. Some related target characterizing experiments were done in a low background shielding house of 8 ton called DÖME, which is able to accommodate the whole detector and sample in its interior. The background level in DÖME is 1 cps in the 0-2 MeV acquisition range.

The LeGe gamma-detector efficiency was determined with certified ^{133}Ba, ^{152}Eu and ^{241}Am calibration sources at 22.7 cm distance. The result is shown in Fig. 1. The relative uncertainty is 1.5% at 40 keV and less than 1% above 90 keV.

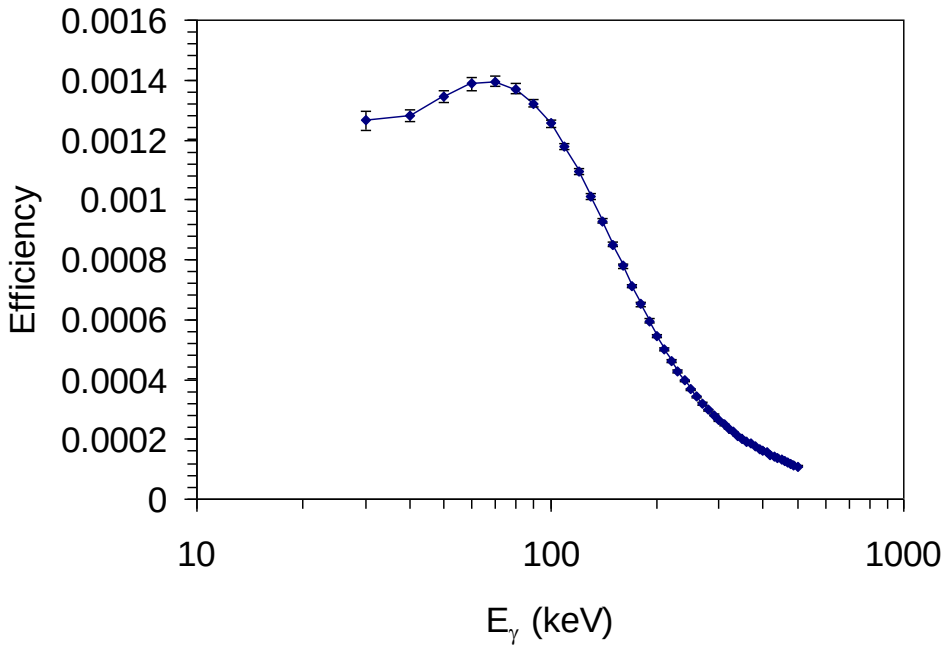

Fig. 1. Efficiency of the LeGe detector.

3. TARGET CHARACTERIZATION AND EXPERIMENTS

The ^{241}Am target is from a 1.27 cm wide, long metal strip existed in the inventory of University of Debrecen. A piece of about 1 cm length was cut. The total mass measured to be 236.4±0.5 mg. Americium-241 metal was sintered on a 0.196 mm thick Ag backing and it was covered with a 2.33-μm thick gold layer. The thickness of the silver backing was determined from attenuation of the 60-keV gamma ray arising from the ^{241}Am layer. The thickness of the gold covering layer was obtained from its activity after the irradiation using the activity of the 25 μm thick gold monitor of a mass of 62.02 mg and taking into account the flux attenuation. The gold monitor foil having a shape in a close match to the target's silver backing separated by a thin Al foil. Finally, they were enclosed in a thin aluminum foil. Most importantly the mass of the ^{241}Am was measured also by utilizing the 60-keV gamma ray intensity, which yielded 9.27±1.8% MBq ≈ 73.1 μg. In this later measurement, the dead time of the counting system was 17.8% which was determined from the reported incoming (ICR) and outgoing (OCR) rates of the XIA PIXE-4 digital analyzer [http://www.xia.com/Manuals/Pixie4_UserManual.pdf].

The target was irradiated at the end of the tapered supermirror guide of the in-beam Mössbauer station [17] of the BRR, facing with its gold layer to the incoming cold neutron beam. The irradiation time was 90120 s.

The decay of the activated monitor foil and the target were measured with the LeGe detector shielded with lead blocks to decrease the room background. The target to detector distance was 22.7 cm to decrease the pile up effect due to the strong gamma and X-rays coming from the ^{241}Am natural alpha decay.

The gold monitor foil was measured for 26847 s after a waiting time of 11.7 days. The dead time of the counting system was negligible.

The decay of the activated ^{241}Am target was followed for 97 hours after a waiting time of 66308 s and saved every hour once. The resulting spectra for the first and the 20th hours are shown in Fig 2.

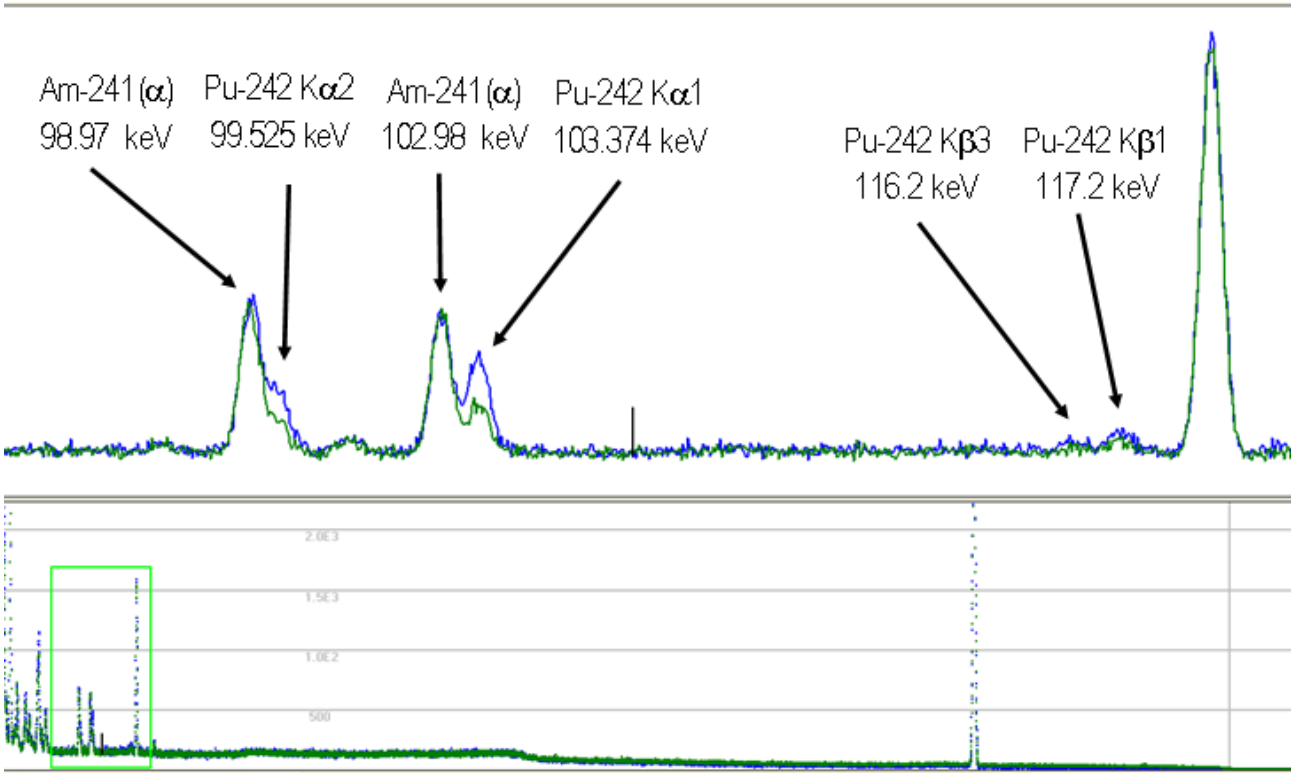

Fig. 2. Portions of measured spectra of the activated ^{241}Am target. The frame in the lower panel is enlarged in the upper panel.

4. DATA EVALUATION

The evaluation of the peak areas between 95-105 keV was done first using Hypermet PC [18, 19] peak-shape fitting program, however it did not provide consistent peak areas for the peak doublets. It was observed that the peak areas arising from the ^{241}Am alpha decay were not constant as a function of time as it would have been expected. It turned out that gamma-ray-X-ray doublets cannot be fitted consistently due to the different peak shapes and peak widths. The X-rays in Pu are wider than the gamma ray peak widths [20].

A different approach was applied to obtain peak areas for the doublets, which is outlined here. First, linear background was determined for each of the 97 spectra, which was subtracted from each channel, then the area of the peak doublets $T_{doublet}$ were calculated by simple summation. In the last step a constant area T_γ plus an area with exponential decay corresponds to the half-life of 16.02(2) h [21] of the ^{242}Am ground state was fit to each one-hour long spectra. The corresponding formula is

$$T(n)_{doublet} = T_\gamma + A_{0\,X-ray} \int_{(n-1)\Delta t}^{n\Delta t} \exp(-\lambda t)dt \qquad n = 1,2,\cdots 97 \tag{1}$$

where $A_{0\,X\text{-}ray}$ is the initial rate of the X-ray peak, n is the serial number of the spectra and Δt is 1 hour.

$$T(n)_{doublet} = T_{Am-241} + \frac{A_{0\,X-ray}}{\lambda} \exp(-\lambda(n-1)\Delta t)\left[1-\exp(-\lambda\Delta t)\right] \tag{2}$$

The peak areas in the doublets were corrected for dead time of the acquisition system before the fit for each value of n. The dead time correction was taken as the ratio (ICR) and (OCR) values of XIA PIXIE-4. The correction values varied between 16.8%-17.8% during the experiment due to the decaying ^{242}Am and ^{198}Au components of the sample. Here the $A_{0\ X\text{-}ray}/\lambda$ is equal to the area that would be obtained from an infinitely long acquisition after the cooling of the sample. The resulting

fits are shown in Fig 3 and Fig 4.

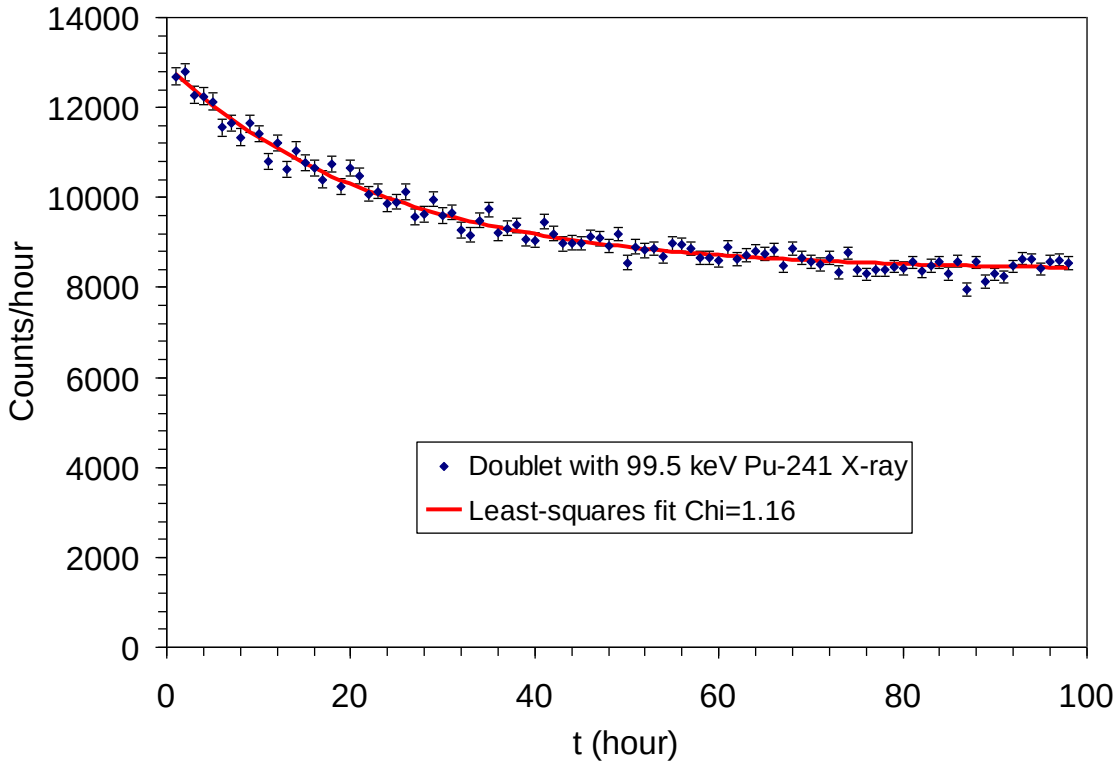

Fig. 3. Fit result for the peak doublet with 99.5 keV Pu-241 X-ray

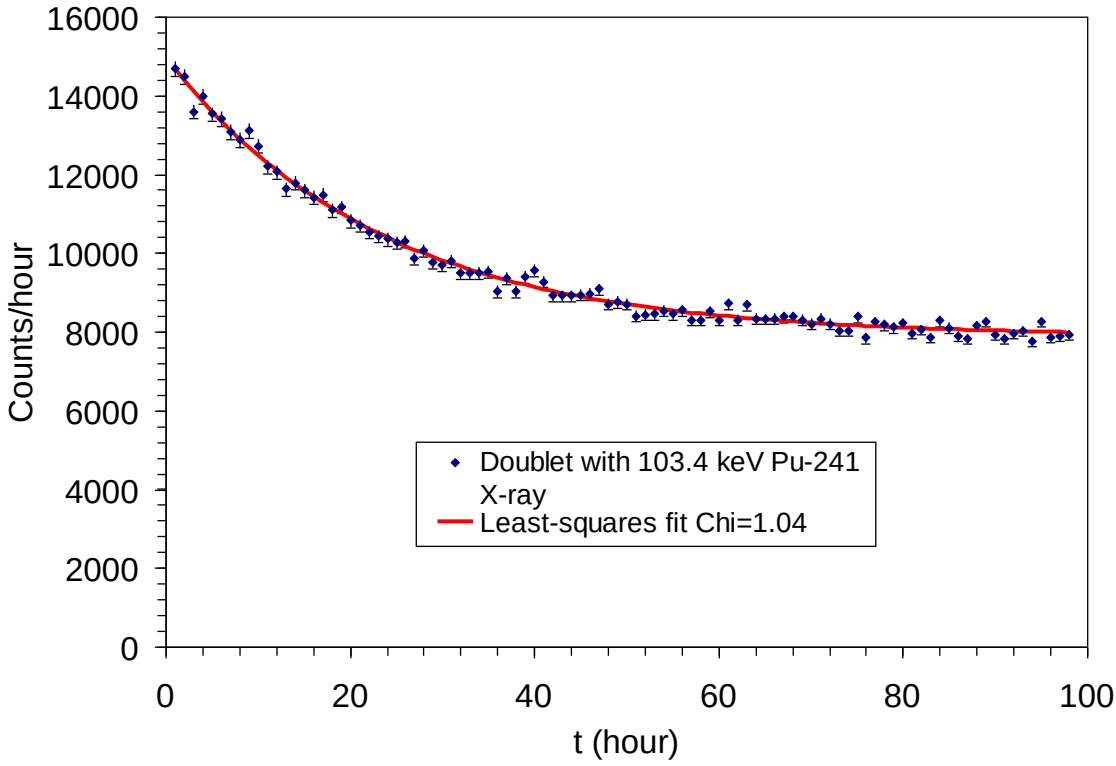

Fig. 4. Fit result for the peak doublet with 103.4 keV Pu-241 X-ray

Both of the fits look excellent using the literature value of 16.02(2) h half-life [21] for the ^{242}Am ground state decay. The nuclear constants shown in Table 2 are obtained from Ref. [22]. The number of ^{242g}Am crated during the activation was calculated from the usual activation formula

$$N_{Am-242g} = \frac{A_{0\,X-ray}/\lambda}{P_{X-ray}\varepsilon \frac{1-e^{-\lambda t_a}}{\lambda t_a} e^{-\lambda t_c}\left(1-e^{-\lambda t_m}\right)}. \quad (3)$$

The $N_{Am\text{-}242g}$ is the number of ^{242g}Am atoms created in the neutron capture during the activation time t_a, $A_{0\ X\text{-}ray}/\lambda$ is the result of the fit above and belongs to infinite acquisition time (t_m=infinite) starting after the cooling period t_c. The symbol λ is the decay constant of the ^{242g}Am and ε is the detection efficiency of the X-ray of ^{242}Pu, which has a production probability of $P_{X\text{-}ray}$.

The evaluation of the number of ^{198}Au created in the activation was calculated from an equation similar to Eq. (3) but using the gold monitor data. This later evaluation yielded 1.24711×10^{12} ^{198}Au atoms with 1.2% uncertainty.

Table 2. Fit result for the two most intense ^{242}Pu X-rays followed the ^{242}Am electron conversion and their absolute production probabilities

$E_{X\text{-}ray}$ (keV)	$A_{0\ X\text{-}ray}/\lambda$	T_γ	χ	$P_{X\text{-}ray}$ ^{242}Pu
99.5	103318±1857	8364±23	1.16	0.0355±0.0017
103.4	160623±1692	7879±21	1.04	0.056±0.003

5. EXTRACTION OF THERMAL NEUTRON CAPTURE CROSS SECTION

Due to the strongly inhomogeneous target, we could not use the traditional comparator method because of the strong absorption of the Ag backing. Instead, we applied a series of MCNP5 calculations, in which we modeled the target as described in section 3. The cold neutron wavelength distribution was measured at the Mössbauer irradiation facility by J. Füzy in pinhole geometry.

The neutron wavelength distribution $\frac{dN(\lambda)}{d\lambda}$ was transformed to neutron energy distribution using the following formulas

$$E(\text{meV}) = 81.92\cdot\lambda^{-2}(\text{AA}) \quad (4)$$

and

$$\frac{dN(E)}{dE} = -\frac{dN(\lambda)}{d\lambda}\left(2\cdot81.92\cdot\lambda^3\right) \quad (5)$$

The normalized neutron energy distributions are shown in Figure 5 for our PGAA and for the Mössbauer station where the ^{241}Am sample was irradiated. The dips in the PGAA distribution are due to monochromators placed upstream in the beam. The PGAA wavelength distribution was measured by us also in pinhole geometry using a position sensitive, ^{3}He filled multi-wire ionization chamber (borrowed from J. Füzy) in a TOF experiment.

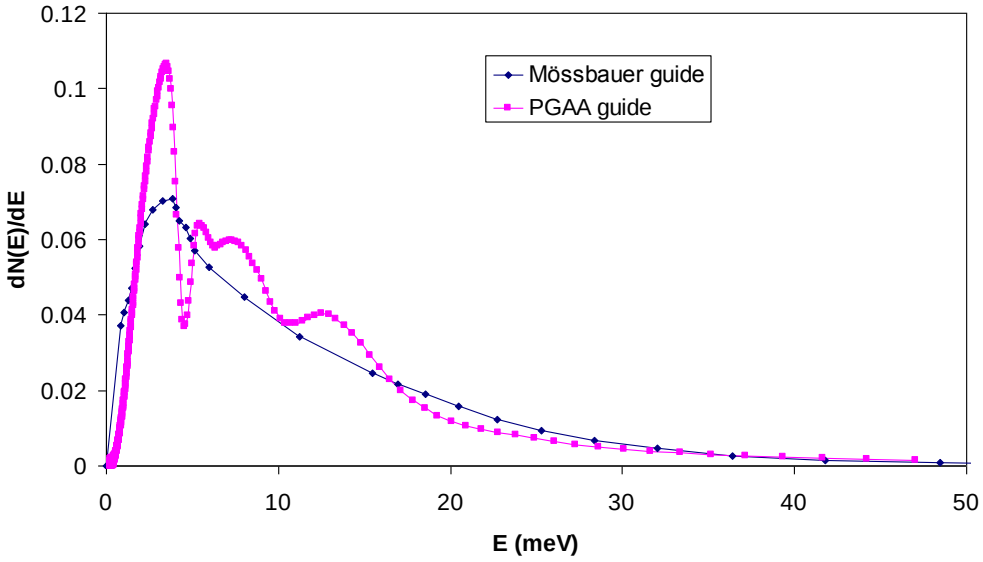

Fig. 5. Normalized neutron energy distributions for the PGAA and the Mössbauer stations

The neutron spectrum of the Mössbauer station was listed in the MCNP5 input file and the neutrons were uniformly sampled in the same area that characterized the sample and started perpendicular to the normal of the sample surface. In the MCNP5 calculations, we used the ENDF/B-VI library values from which cross section values at 25.3 meV are listed in Table 3 for ^{241}Am.

Table 3. ENDF/B-VI Cross section library values for ^{241}Am

Type	Cross section (b)	MT number
(n,γ)	618.6	102
(n,tot)	633.0	1
(n,el)	11.3	2
(n,f)	3.2	18

F4 tallies with Fm4 modifier were calculated for the layers of the sample to obtain number of atoms created in radiative capture for one started neutron. This means the simulation of quantity

$$\frac{n}{V}\int \sigma_{capture}(E)\phi(\vec{r},E)dEdV \qquad (6)$$

where n/V is the atom density in the cell (layer), $\sigma_{capture}(E)$ is the capture cross section, ϕ is the scalar flux.

The thickness of the covering gold layer was changed until it provided the same number of starting neutrons as the measured one calculated from the activity of the monitor foil and the MCNP5 simulation. However, this change influenced the absorption of the ^{241}Am 60 keV gamma ray and the number of ^{242}Am induced ^{242}Pu X-rays, which resulted in the change of the ^{241}Am mass and the number of the ^{242}Am produced. One iteration with a new value for the thickness of the covering gold layer was sufficient to achieve self-consistent result.

The thermal capture cross section of ^{241}Am was obtained from the comparison of the measured ^{242}Am atom number created in the irradiation and the same number calculated with the MCNP5 using the starting number of particles determined with the gold monitor foil. The necessary number of starting particle (SP) in the MCNP5 modeling to produce the number of ^{198}Au measured value was calculated to be SP=5.25497×10^{13}. Since the measurement (using the 103.4 keV X-ray) yielded only the ground state production, the measured ^{242g}Am production was scaled up using the ground state branching ratio of 0.914 to obtain the total ($^{242(g+m)}$Am) capture production of 9.51346×10^{9}.

The gold monitor SP value was used to divide the measured production value (9.51346×10^{9}) of the $^{242(g+m)}$Am to calculate the production value for one staring particle, which yielded 1.81037×10^{-4}. The same value from the MCNP5 calculation was 1.8888×10^{-4}, indicating that the cross section in the ENDF/B-VI is larger by a factor of 1.0433 thus the cross section according to our measurement should be 592.9 b=618.6 b/1.0433 at the 25.3 meV energy. The same method was applied for the 99.5 keV X-ray, which yielded 588.5 b for the same cross section.

The relative uncertainty calculation has the following statistical and systematic components for the ^{241}Am measurement, which is summarized in Table 4. The same is summarized for the ^{197}Au monitor foil measurement in Table 5.

Table 4. Uncertainties for the 241Am capture measurement

$E_{X\text{-}ray}$ (keV)	99.5		103.4		Comment
Uncertainty	Statistical	Systematic	Statistical	Systematic	
$A_{0\ X\text{-}ray}/\lambda$	1.8%	0%	1.0%	0%	From fit
ε(E)	1.0%	0.5%	1.0%	0.5%	Syst: Geometry
# of ^{241}Am	1.4%	1.1%	1.4%	1.1%	
$P_{X\text{-}ray}$	0%	4.8%	0%	5.4%	
λ of ^{242g}Am	0%	0.1%	0%	0.1%	
$N_{Am\text{-}242g}$	2.1%	4.8%	1.4%	5.4%	
Branch g.s.	0%	0.8%	0%	0.8%	Fioni et al. [4]
$N_{Am\text{-}242(g+m)}$	2.1%	4.9%	1.4%	5.5%	
# of ^{242}Am per starting particle	1.4%	1.1%	1.4%	1.1%	MCNP value

Table 5. Uncertainties for the ^{197}Au monitor foil capture measurement

E_γ (keV)	411		Comment
Uncertainty	Statistical	Systematic	
A_γ	0.1%	0%	From fit
ε(E)	1.0%	0.5%	Syst: Geometry
# of ^{197}Au	0.1%	0%	
P_γ	0%	0.1%	
λ of ^{198}Au	0%	0.1%	
$N_{Au\text{-}198}$	1.0%	0.6%	
# of ^{198}Au per starting particle	0.1%	0%	MCNP value

Using the relative uncertainties from Table 4 and we deduced a value of 592.9 b ±(2.2% statistical and 5.6% systematic, total 6.0%) for the ^{241}Am thermal (2200 m/s) capture cross section yielding to the ground and metastable state of ^{242}Am from the 103.4 keV ^{242}Pu X-ray. From the 99.5 keV X-ray it is 588.5 b ±(2.7% statistical and 5.1% systematic, total 5.8%). The un-weighted average is 591 ± 35 b. The ground state cross section is 540 ± 32 b, which is at the low end of the values presented in Table 1.

6. CONCLUSIONS

The ground state cross section of ^{241}Am has been measured with a beam of cold neutrons using the X-ray emission of the decay product of ^{242}Pu. This methodology avoids the uncertainty caused by resonance neutrons in the pile activations.

The mean value of 591 ± 35 b of our total capture cross section for ^{241}Am is at the low end of the most recent values, but is in agreement with most of them within its uncertainty. The uncertainty is mainly coming from the systematic uncertainty of the ^{242}Pu X-ray production probability in the electron conversion of the ^{242}Am ground state.

7. ACKNOWLEDGEMENTS

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