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Neutron transmission and capture of ^{241}Am

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Abstract. A set of neutron transmission and capture experiments based on the Time Of Flight (TOF) technique, were performed in order to determine the ^{241}Am capture cross section in the energy range from 0.01 eV to 1 keV. The GELINA facility of the Institute for Reference Materials and Measurements (IRMM) served as the neutron source. A pair of C_6D_6 liquid scintillators was used to register the prompt gamma rays emerging from the americium sample, while a Li-glass detector was used in the transmission setup. Results from the capture and transmission data acquired are consistent with each other, but appear to be inconsistent with the evaluated data files. Resonance parameters have been derived for the data up to the energy of 100 eV.

1 Introduction

New reactor designs created a need for accurate cross sections for neutron induced reactions for the major and minor actinides [1]. The ^{241}Am is considered as the most important because of its properties in terms of activity, half-life and mainly radio-toxicity. Deep geological isolation is not a favorable solution and a more sophisticated approach should be followed with incineration and/or transmutation being considered as viable options.
During the last decade four new measurements [2-5] have been devoted to determine the thermal capture cross section of ^{241}Am. The values of these four measurements are in agreement with each other, but basically disagree with the evaluated data files, except for JEFF-3.1, which has already taken them into account. Before the measurement of Jandel et al. [2] only a handful of experimental values were available: one capture measurement by Weston et al. [7], and three transmission measurements reported by Kalebin et al. [8], Adamchuk et al. [9], and Slaughter et al. [10]. The results of all these measurements do not agree with each other. In addition the resonance integral determined by the resonance parameters was not in agreement with experimentally observed ones. The latest measurement by Jandel et al. [2] reported resonance parameters with total width smaller by a few percent. The ratio between the neutron widths reported by Jandel et al. and [7], [8] and [11], as depicted in figure 1, shows an energy dependence. Therefore another set of independent experiments would be desirable for a reliable new evaluation.

2 Experimental details

The experiments were performed at GELINA (GEel LINear Accelerator) [12], a pulsed white neutron source at the Institute for Reference Materials and Measurements in Geel. GELINA is a linac

designed and built for high resolution cross section measurements. Intense pulsed electron beams with repetition rates ranging from 10 to 800 Hz are accelerated up to an energy of 150 MeV with a peak current of 12 A. Using a post-acceleration compression magnet, the width of the electron pulses can be reduced to less than 1 ns. After the compression, these high-energy electrons hit a mercury cooled uranium target. In the target the electrons produce Bremsstrahlung, and subsequently neutrons via (γ,n) and (γ,f) reactions. Two water filled beryllium containers mounted above and below the neutron production target, are used to moderate the neutrons. Applying different neutron beam collimation conditions, experiments can use either a fast or a thermalized neutron spectrum.

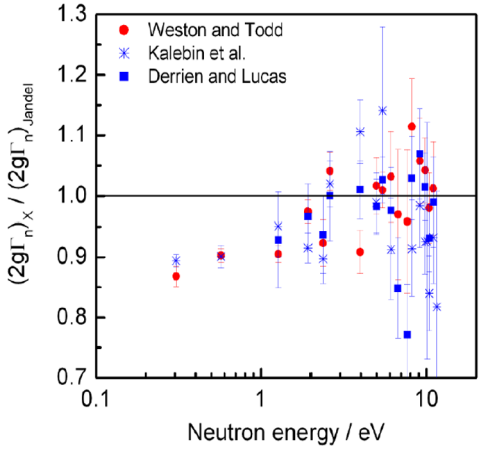

Fig. 1. Ratio of Γ_n from previous experiments to that of Jandel et al..

The neutron production rate is constantly monitored by BF_3 counters that are mounted on the ceiling of the target hall. For all experiments reported in the present work, the background was determined using the black resonance technique [13], i.e., filters of appropriate thickness are inserted into the beam to absorb all neutrons at a given energy. Typical filter materials used in these experiments are Ag, Co and Na.

2.1 Americium sample

For this type of experiments, the sample's homogeneity is of utmost importance. To achieve the best possible conditions, a special technique was adopted for the sample preparation. About 3 g of Y_2O_3 was prepared by the sol-gel method in the form of porous beads. This structure of 12 mm diameter, was used as the matrix in which approximately 320 mg of Am was infiltrated. A 0.5 mm window Al canning encapsulated the material. The total ^{241}Am content in the sample has been determined by calorimetry measurements at JRC-ISPRA and been compared in very good agreement with gamma spectroscopy results obtained at the IRMM [14].

2.2 Transmission setup

The transmission measurements have been performed at flight path 2 of GELINA, a flight path that views the moderator at an angle of 9 degrees with its normal. The chosen flight path length was 26.45 m. The beam diameter was limited to approximately 2 cm, using a combination of lithium-carbonate, copper and nickel collimators. The sample was positioned at a distance of approximately

10 m from the neutron source, so halfway between the neutron target and the detector system. For neutron detection a 1.27 cm thick lithium-glass scintillator was employed, with a diameter of approximately 10 cm viewed by two EMI 9823 QKB photomultiplier tubes.
To reduce the impact of a variation of the neutron flux on the determined transmission, measurements of the sample-in and sample-out of the beam were cycled every 20 minutes. The time spectra of the BF_3 counters mentioned earlier, were recorded in parallel and used to normalize these acquired data to the same total neutron fluence.

2.3 Capture setup

The experiments for determining the capture yield of americium were performed at a flight path length of 10 m with the beam diameter at the sample station reduced to approximately 75 mm using lithium-carbonate plus resin, copper and lead collimators. The detection of the photons that were produced in the capture event is accomplished by two cylindrical C_6D_6 detectors (NE230). These detectors are mounted at an angle of 125 degrees to reduce the effects caused by the anisotropy of the dipole radiation. The pulse height weighing technique was used to create a detector response proportional to the energy of the registered gamma. A description of the method and of the calculation of the weighting function can be found in Borella et al. [15]. The incident neutron flux was recorded in parallel using a ^{10}B ionization chamber. The chamber is placed approximately at a distance of 1 m prior to the capture detection system. The $^{10}B(n,\gamma)$ standard reaction is used to determine the energy dependence of the neutron flux below the keV region.

3 Data reduction and analysis

To derive the transmission factor and capture yield from the TOF spectra, the software package AGS (Analysis of Geel Spectra) [16] was used. Within its operations it performs the most important actions, such as dead-time correction, background subtraction etc., while it fully propagates uncertainties, starting from counting statistics.
The transmission factor $T(t_n)$, as a function of the neutron time-of-flight t_n, was obtained as the ratio of the time-integrated spectra of the sample in and out, C_{in} and C_{out}, after been corrected for the incident neutron flux and the dead-time effects. The shape of background was determined from fitting the black resonance dip points in the TOF spectrum.
During the capture measurement, data were stored in cycles of 1 hour, both for the scintillators and the flux chamber. The background of the neutron flux was thoroughly studied applying the black resonance technique. A set of sulfur, sodium, cobalt and tungsten filters of appropriate thickness, were used to absorb all neutrons at their resonance 102 keV, 2.8 keV, 132 eV and 18.8 eV respectively. It should be noted that also in the case of capture a filter of Na placed in the beam served to control the level of background.
For obtaining reliable values from the capture measurement the determination of the optimum threshold is very important. The choice of threshold will strongly determine the signal to background level - and therefore finally the accuracy of the experiment. For a non-radioactive sample a proper threshold can be as low as 100 keV, a value below the energy of most gamma rays emitted in the cascade. In the case of radioactive isotopes a higher limit as 300 keV can be sufficient. For the americium used in our experiments the best signal to background was obtained for a threshold of 650 keV (figure 2).
Using all the above information, the experimental observables (transmission factor and capture yield) were obtained.

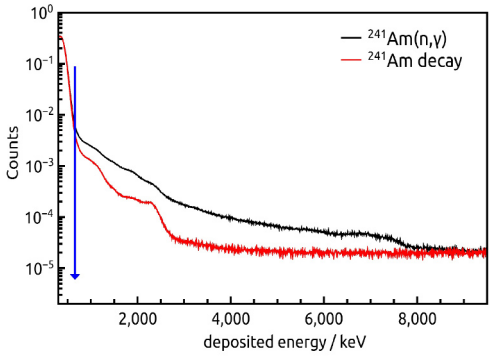

Fig. 2. Amplitude spectrum indicating the threshold used corresponding to 650 keV of deposited energy.

4 Results and discussion

Comparing the total observed widths, the Doppler broadening and the intrinsic widths, one can conclude that a resonance shape analysis - and therefore a determination of E_{res} , Γ_n and Γ_γ is only feasible for low energies. The fitting process was done with the resonance shape analysis code REFIT [17].

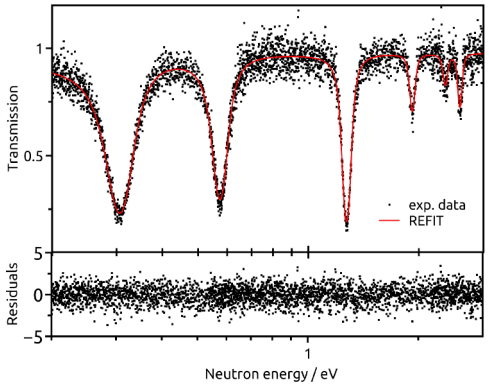

Fig. 3. Experimental points from the trasmission of ^{241}Am along with the fitting curve.

The normalization factor was determined by the fit of the transmission data, using the extracted resonance parameters of the first three resonances. The factor obtained agreed within 1% in all three cases. Figures 3 and 4 illustrate the the experimental data points from the two experiments along with the fitting curves obtained from the REFIT code and their residuals. Extracted resonance parameters for the first three resonances are in relative agreement with the recent experimental

results. For higher energies the resonance parameters appear to be larger. The same stands for the thermal point cross section. The final analysis will clarify the exact level of difference.

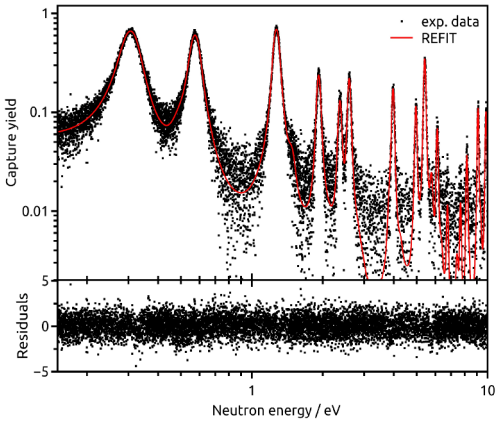

Fig. 4. The americium capture data points along with the fitting curve from REFIT.

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