MEASUREMENTS OF NEUTRON CAPTURE CROSS SECTIONS RELEVANT FOR NUCLEAR WASTE TRANSMUTATION, BY γ AND α SPECTROSCOPY

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We have performed neutron irradiations at the High Flux Reactor of the Institut Laue-Langevin (Grenoble) on ^{243}Am and ^{242}Pu targets. Using γ and α spectroscopy of irradiated samples, we have measured Maxwell averaged neutron capture cross sections, respectively (74.8 ± 3.3) b and (22.7 ± 1.1) b, in a 98% thermal neutron spectrum at $T \simeq 50°C$.

1. Introduction

One of the crucial challenges for the nuclear industry in this century will be its capability to manage and reduce the inventory in mass and the radiotoxicity of the nuclear waste coming from the spent fuel of nuclear power plants. One of the solutions presently under study by the scientific community is the transmutation of long lived isotopes into shorter lifetime or stable isotopes. The realization of nuclear waste incinerators requires - in addition to innovative technological solutions necessary to built systems working at very high neutron fluxes ($> 10^{15}$ $n/s/cm^2$) - a reliable set of nuclear data, such as neutron capture and fission cross sections, for all the isotopes involved in the transmutation chain and for a wide range of neutron energy, covering thermal to fast spectra. Minor actinides, due to the role they play in conventional fuel cycles, have been little studied during the past decades. Their nuclear parameters tabulated in the three most widely used evaluated nuclear data libraries (ENDF-B6 [1], JEF-2.2 [2] and JENDL-3.2 [3]) are not always known with the precision required for transmutation dedicated systems

In this context, following the measurement of the $^{242gs}Am$ capture cross section [4] in order to confirm the theoretical possibility to transmute ^{241}Am in a high thermal neutron flux, the Mini-Inca project at *the Direction des Sciences de la Matiere* of CEA/Saclay, has widened the field of investigations to all other minor actinides which play an important role in the nuclear waste inventory. Thus, between 2000 and 2001, two experiments have been performed to measure ^{243}Am and ^{242}Pu neutron capture cross sections [5].

2. Experimental method

The experiments have been carried out at the High Flux Reactor of the Institut Laue-Langevin of Grenoble, where ^{243}Am and ^{242}Pu samples have been irradiated in the H9 neutron beam. The main characteristic of H9 is to provide a 98% thermal neutron flux with a very high intensity of 6.10^{14} $n/s/cm^2$. After irradiation, samples are automatically transfered to the "Mini-Inca" chamber where γ and α spectroscopy are performed to measure appropriate γ or α lines counting rates corresponding to the isotopes formed during the irradiation. From the $A_{\gamma,\alpha}$ line intensities, one can extract the amount N_{I_f} of isotope I_f produced:

$$N_{I_f} = \frac{A_{\gamma,\alpha} \times \tau}{\Omega\epsilon} \exp\left(\frac{t}{\tau}\right), \tag{1}$$

where τ is the life time of isotope I_f, t is the cooling time and $\Omega\epsilon$ the detection efficiency. A comparison with the measurement of remaining initial isotope N_{I_i} gives the capture cross section[a]:

$$\sigma_c(I_i) \simeq \frac{1}{\Phi.t_{irr}}.\ln\left(1 + \frac{N_{I_f}}{N_{I_i}}\right) \tag{2}$$

where t_{irr} is the time of irradiation at the integrated neutron flux Φ.

2.1. *The Mini-Inca chamber and experimental setup*

The vacuum chamber is mechanically coupled to the H9 target exchanger[6]. A movable rail drives the sample holder to a calibrated position in front of a γ and α detectors.

The sample holder is a 0.4 mm thick titanium frame (see figure 1), where the actinide sample and a flux monitor are placed. The mass of the active deposit is chosen to be lower than 20 μg to limit the α, γ-ray self absorption and the radioactivity of irradiated samples.

[a] Assuming I_i fission cross section and I_f burn-up negligible

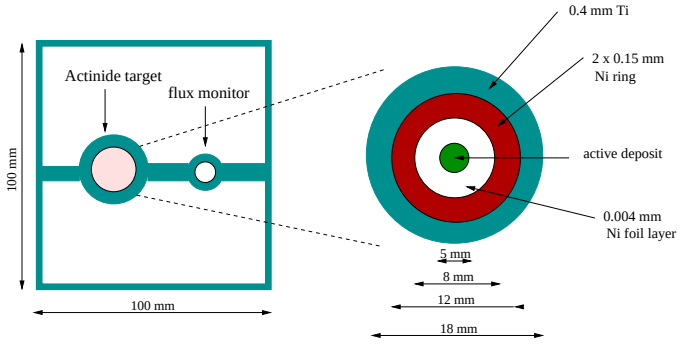

Figure 1. Mini-Inca sample holder and target layer.

The γ detector is a coaxial Germanium ($Diameter = 5.15\ cm$, $Thickness = 4\ cm$) with a 500 μm carbon-epoxy window. A transistor reset preamplifier coupled to a Digital Signal Processor Unit allows high counting rates acquisition ($\sim 80\ KHz$) with dead time lower than 60%. The detector is placed on a movable rail allowing a positioning from 40 cm to 80 cm with respect to the sample, leading to detection solid angles : $2.6\ 10^{-4} \leq \Omega/4\pi \leq 1.10^{-3}$.

The α detector is a Passivated Implanted Planar Silicon ($Diameter = 1.13\ cm$, $Thickness = 300\ \mu m$) placed also on a movable rail that allows a positioning from 1 cm to 30 cm with respect to the sample, leading to detection solid angles : $1.10^{-4} \leq \Omega/4\pi \leq 5.10^{-2}$. A 3 mm thick steel plate collimator is placed in front of the target to reduce the amount of β particle background in the detector. The typical detector resolution under experimental conditions is 20 KeV.

The Germanium photo-peak efficiency has been determined experimentally with calibrated standard gamma sources (^{152}Eu, ^{241}Am and ^{60}Co). Results are plotted in figure 2 and reveal an efficiency of $\epsilon = 0.098 \pm 0.002$ at 1.17 MeV.

2.2. *Neutron flux measurement*

The average neutron flux intensity is measured with a monitor irradiated together with the actinide sample as shown in fig 1. It consists of a 6 mm diameter Al foil doped with 1% Co. The mass of the monitor is typically 8 mg and the homogeneity is 2%. Knowing the ^{59}Co Maxwell average neutron capture cross section (31.4 ± 0.6) b[7] and the photo-peak efficiency for ^{60}Co γ line energies, the flux intensity has been found to be $\Phi = 6.10^{14}\ n/s/cm^2$ with a relative uncertainty of 3.8%, for both experiments.

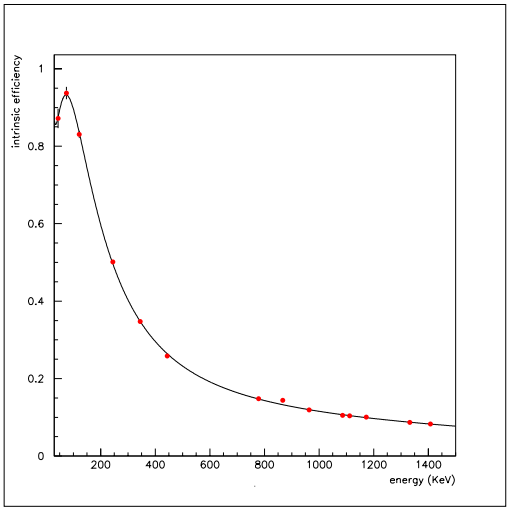

Figure 2. Photo-peak efficiency of the Germanium as a function of the photon energy. Dots are obtained from standards and the line is a polynomial fit.

3. Measurement of $^{243}Am(n,\gamma)^{244}Am$

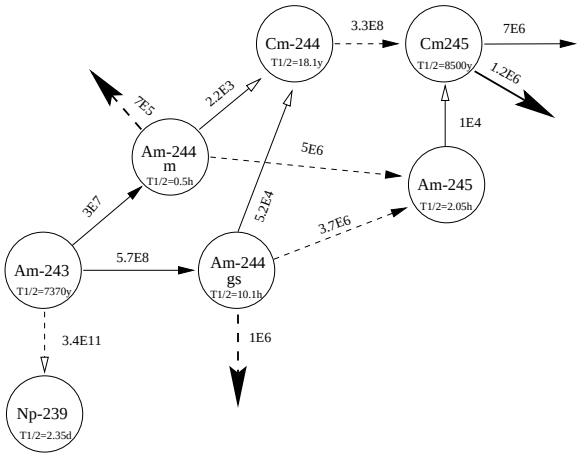

Figure 3. Schematic view of the isotopes produced in the ^{243}Am chain

The cross sections are determined from the amount of ^{244}Am formed, after the irradiation of ^{243}Am. As the half-life of ^{244m}Am is 26 mn, only

$^{244gs}Am$ ($T_{1/2}$ = 10.1 h) is measurable (with the 744 KeV γ line) after several hours of cooling time. As shown in figure 3, the ^{243}Am total capture cross section can be determined via a counting of ^{244}Cm (α lines at 5805 and 5763 KeV), after complete decay of the two states of ^{244}Am. We have thus irradiated 11.7 μg of ^{243}Am during 3.5 h. The γ and α spectra obtained after irradiation are presented in figure 4. Due to the presence of an external α contamination in H9, we have adjusted an empirical shape for all the α peaks to subtract background and to account properly for pile-up and resolution degradation due to electronic effects.

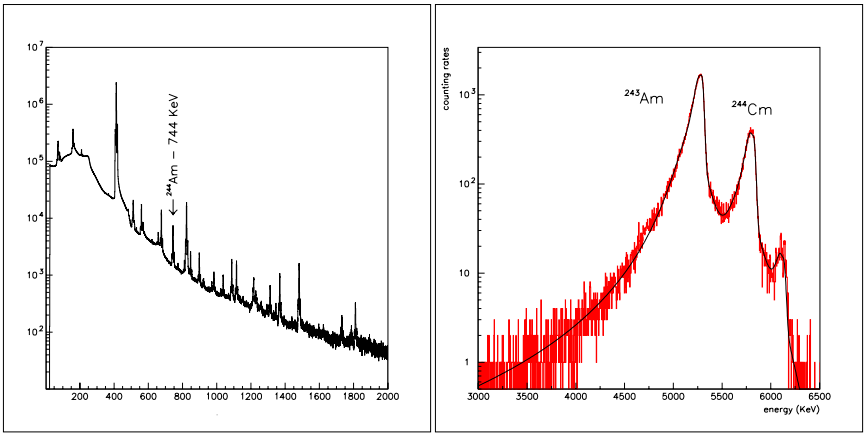

Figure 4. γ (left) and α (right) spectra of ^{243}Am irradiated sample after respectively 17 h and 6 d of cooling.

Using equation 2, we have extracted ^{243}Am thermal neutron capture cross sections. The σ_0 values at 0.025 eV are deduced from the measured 50°C Maxwell averaged values, with the hypothesis of a $\frac{1}{v}$ behavior (g=1.013) of the cross section. The results are presented in table 1:

Table 1. ^{243}Am thermal neutron capture cross sections

Cross section	$< \sigma >_{50°C}$ [b]	σ_0 **[b]**	relative uncertainty [%]
$\sigma_c^{tot}(^{243}Am)$	74.8	**87.5**	4.4
$^{243}Am(n,\gamma)^{244gs}Am$	4.7	**5.5**	30.1

$^{243}Am(n,\gamma)^{244gs}Am$ cross section is compatible[b] with previous experi-

[b]The 30% uncertainty comes mainly from the 744 KeV γ line intensity [7]

mental data [8,9], whereas $\sigma_c^{tot}(^{243}Am)$ is found 11% higher than the adopted values in the commonly used data libraries[3,2,1] (76 ± 1) b. On the other hand, this measurement is compatible with the most recent previous experimental data[10] (83 ± 6) b.

4. Measurement of $^{242}Pu(n,\gamma)^{243}Pu$

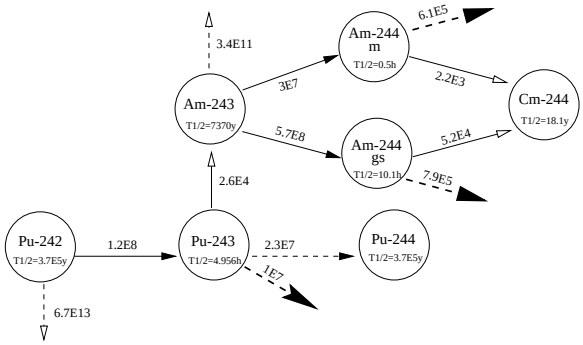

Figure 5. Schematic view of the isotopes produced in the ^{242}Pu chain

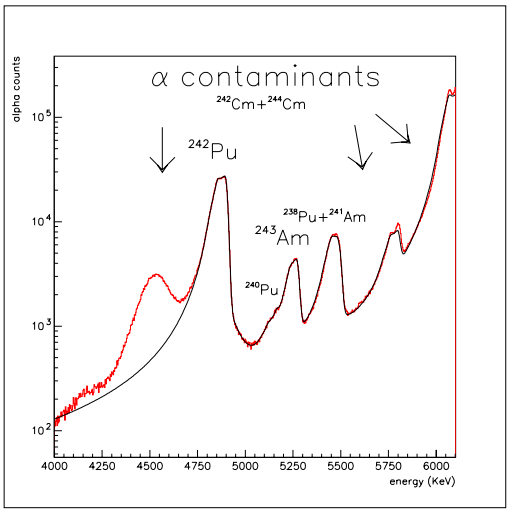

Figure 6. α spectrum of ^{242}Pu sample after 2 d of irradiation. Note the broad α peak at low energy corresponding to H9 contaminants deposited in the back side of the target layer.

In this measurement, cross sections are determined from the amount of ^{243}Am formed after the complete decay[c] of ^{243}Pu. We have irradiated 10.5 μg of ^{242}Pu during 2 d. From figure 6 and equation 2, we have measured the $50°C$ Maxwell averaged neutron capture cross section:

$$< \sigma_c(^{242}Pu) >_{50°C} = (22.7 \pm 1.1)\ b$$

and deduced $\sigma_0 = (26.6 \pm 1.3)\ b$ at 0.025 eV. This value is 40% higher than adopted evaluated values. Only one over 6 experiments performed between 1954 and 1979 gives a compatible value [11].

5. Conclusions

Using γ and α spectroscopy of irradiated samples, we have measured ^{243}Am and ^{242}Pu neutron capture cross sections at thermal energy [5]. These results differ slightly from previous existing data but lean on a simple and reliable analysis, due to the high level of neutron thermalization available at the HFR of ILL,. In the future, we will extend our experimental program to other minor actinides of interest [12] (^{237}Np and ^{243}Cm) in order to improve nuclear data necessary for the study of the optimal incineration conditions for the next generation systems dedicated to the transmutation of the nuclear waste.

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[c]Due to a high γ background, the measurement of the 84 KeV line associated with the β decay of ^{243}Pu, was not possible