Measurements of the β-decay half-lives and γ-ray emission probabilities of ^{244gs}Am and ^{244m}Am isotopes for the ^{243}Am capture cross sections determination.

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Abstract

Recently it was required by an IAEA Coordinated Research Project further studies on the decay of actinides and in particular on the β-decay of ^{244}Am. In this paper we report on an experiment to measure the β-decay half-lives of the isomeric and the ground states of ^{244}Am. These data were obtained thanks to the use of ADONIS a high counting rate acquisition system. The obtained values amount to $T_{1/2}$=28.3±1.3 minutes for ^{244m}Am and $T_{1/2}$=10.65±0.12 hours for ^{244gs}Am that is slightly higher than the evaluated ones. Thanks to the use of coupled α- and γ-spectroscopic techniques we also measured the total thermal capture cross section and the capture branching ratio to the isomeric state of ^{244}Am which amount to $\sigma_{tot} = (81.3 \pm 2.5)$ and $BR = 0.953 \pm 0.02$ respectively. Using these measured partial cross sections we have then calculated new absolute γ-emission probabilities for the decay of the ground and the isomeric states. No large discrepancies were observed with previous evaluated data, but we largely reduce the uncertainties on the intensities.

Keywords: Thermal Cross section, Decay Data, Isomeric state, Half-life, Activation technique, Transmutation, Nuclear waste

1. Introduction

The ^{244}Am isotope is an important actinide for high burn-up fuel within the reactor core and for Minor Actinide transmutation within dedicated systems. It is produced by neutron capture on ^{243}Am and quickly decays to ^{244}Cm (see Fig. 1), which largely contributes to the decay heat and radiotoxicity of the fuel and the nuclear wastes. The ground and isomeric states of ^{244}Am present large thermal fission cross sections of about 2000 b but due to their short life high fluxes of thermal neutrons are needed to directly incinerate these isotopes. These neutron fluxes can be provided by future dedicated transmutation systems but the economy of the reaction is strongly driven by the decay half-lives of the ground and isomeric states of ^{244}Am.

The half-life of the ^{244}Am ground state was measured by Vandenbosch and Day (1962) who found a value of (10.1±0.1) h, that is, up to now, the only existing measurement. For the experiment it was used a β-spectrometer in addition to γ-detector to establish the decay scheme of ^{244}Am which was later completed by Hoff et al. (1984) who measured the γ-rays with a high resolution curved-crystal spectrometer and the conversion electrons with a high resolution β-spectrometer. The 1+ isomeric state was observed for the first time by Street et al. (1950) in the neutron irradiation of ^{243}Am target. They found a β-activity with a half-life of 26 min that they attributed

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to an isomeric state. Latter on, Ghiorso et al. (1954) found a value of 25 min for the half-life by using a NaI-crystal spectrometer. The evaluated value by Akovali (2003) is of (26±1) min.

Recently an IAEA coordinated research project (CRP) (Kellet et al. , 2008) dedicated to the decay data library for actinides has required further study to improve the quality of the half-life value for the ground state and measurements to quantify the value and uncertainty with much greater confidence for the isomeric state.

In a previous experiment Marie et al. (2006) we determined the total ^{243}Am(n,γ)^{244}Cm and the partial ^{243}Am(n,γ)^{244gs}Am cross sections for thermal neutrons, using coupled α- and γ-spectroscopic techniques. Unfortunately, in this experiment the input counting rate in the γ-detector was too important for our acquisition system to be able to measure the β-decay of the isomeric state and the branching ratio to this level. Then we repeated the experiment using a high counting rate acquisition system. In this paper we report on the new results obtained in this experiment, on the determination of the half-lives of ^{244gs}Am and ^{244m}Am isotopes and on the neutron capture cross sections of ^{243}Am to the isomeric and ground states of ^{244}Am.

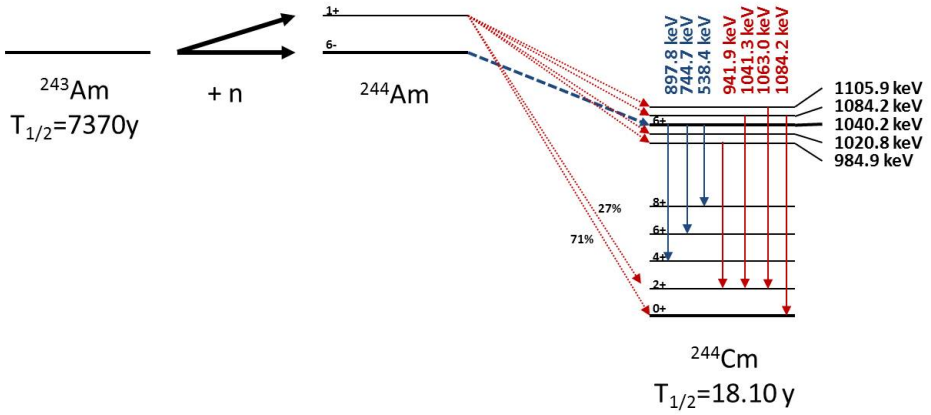

Figure 1: Reaction chain relevant to the ^{243}Am(n,γ)^{244}Cm reaction and simplified β-decay scheme of ^{244}Am.

2. Experimental details

The experiment was performed at the High Flux Reactor of the Laue-Langevin Institute in Grenoble, using the Mini-INCA set-up (Marie et al. , 2006). It is composed by a vacuum chamber which is directly connected to the H9 sample changer allowing to measure the irradiated sample right after the irradiation. A High Purity Germanium (HPGe) detector and a Passivated Implanted Planar Silicon (PIPS) detector are used to measure the γ- and α-emissions of the irradiated samples. These two detectors are placed on movable trolleys, looking at the center of the target. The HPGe detector is a N-type coaxial Ge detector with reset-type preamplifier and thin entrance Be window.

Two irradiations were performed in the H9 channel with the same sample. The first irradiation of (3.277±0.002) hours was to measure the total neutron capture cross section of ^{243}Am, whereas the second one of (24.00±0.02) min was to measure the partial cross sections and the decay properties of the ground and isomeric states of ^{244}Am. About one day of cooling was respected between the two irradiations to performed the measurements. The ^{243}Am sample was electrodeposited onto a 4 μm Ni backing AlMahamid et al. (2005) and characterized before irradiation using the PIPS-detector. We deduced an effective deposited mass of (10.926±0.11) μg of ^{243}Am and a purity of 99.997% (0.003% in mass of ^{238}Pu).

For both irradiations, the sample was transfered to the vacuum chamber for measurement about half an hour after irradiation. Thus when the measurements started, about half of the isomeric state had already decayed into ^{244}Cm. The total transfer time was measured precisely for each irradiation and a special care was adopted for the timing of the second experiment. Indeed, it takes about 6 min for the sample to be moved to the irradiation position and the same time to be removed. During these transfer times, the neutron flux is quasi linearly increasing and decreasing, respectively. These phases were included into our analysis taking into account the time and velocity data given by the operators during the transfer. We measured a total transfer time from the irradiation position to the measurement position of (32.9±0.02) min.

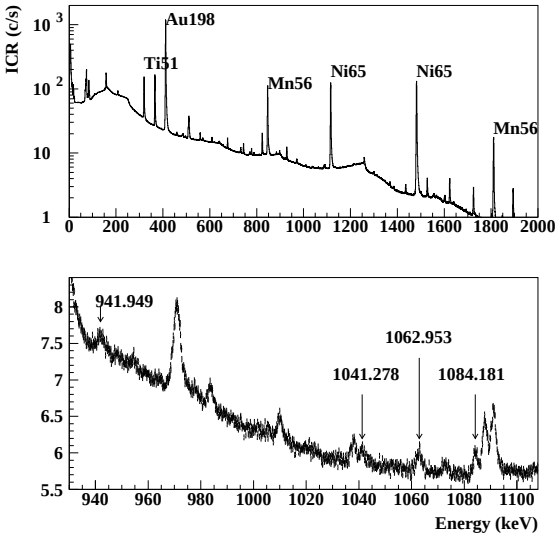

Figure 2: An example of γ-energy spectrum recorded after irradiation (upper panel) where we see the dominant γ-rays due to the activation of pieces supporting the sample. A zoom is done on the energy-region (lower panel) where appear the γ-rays from ^{244m}Am (indicated values).

One of the main challenge in this measurement was the high counting rate right after the irradiation due to the activation of the sample and its surrounding, particularly the Ni backing (see Figure 2). The rate was strongly reduced by the 5 cm thick W collimator placed in front of the HPGe detector but still high enough to saturate even the best commercial high counting rate system developed by Canberra, the Digital Signal Processor (DSP2006). Then we used the ADONIS system (Barat et al. , 2006) developed at CEA/DRT that we already used for similar measurement with short-life isomer (Belier et al. , 2006). The ADONIS system consists in a digital system receiving directly the pulses supplied by the preamplifier, and after a first amplification stage converts the signal with an analog-to-digital converter module operating at 10^7 samples/s (14 bits). Then statistical-based complex correction algorithms are applied to treat the signal, the pile-up of events and the noise. Finally, ADONIS gives an estimation of the input counting rate (ICR) and its variance as a function of the energy, from the counting of the "on" over "off"-periods defined by the charge collection in the detector ("on" period corresponds to a charge collection). This system has been validated up to 1 MHz (Barat et al. , 2006) and shows small bias on the quantitative response that does not exceed 1% up to 300 kHz and less than 4% at 1 MHz. Up to now it is the only system able to operate with such characteristics at this level.

3. Analysis and results

3.1. Decay half-lives

The second irradiation was used to determine the decay half-lives of the ground and isomeric states of ^{244}Am. The half-lives were extracted from the measured evolution as a function of time of their γ-transition activities $a_i(t)$ deduced from the photopeak activities ($ICR(E_i,t)$):

$$a_i(t) = \frac{ICR(E_i,t)}{\Omega\varepsilon} \tag{1}$$

where $\Omega\varepsilon$ is the total efficiency of the photopeak determined with ^{60}Co and ^{152}Eu standard sources[1]. The photopeak activity was fitted with a Gaussian function superimposed to a linear background. Because of the short lifetime of the isomeric state and the fast decrease of the activity, we choose to run the acquisition during all the decay of the isomeric state without stopping it. The ICR estimator was then based on the accumulation of events during all the time of the measurement. The consequence of such integration method is the presence of remnant activities, as seen on Figure 3. An estimator of the partial activities can be constructed as:

$$\widetilde{a}_i(t) = I_\gamma^i A(T_{irr2}) e^{-\lambda_i T} \frac{\int_0^t e^{-\lambda_i u} du}{t} \tag{2}$$

where I_γ^i is the γ-intensity of the γ-ray i and $A(T_{irr2})$ is the activity by the end of the second irradiation, T the time elapsed since the end of the irradiation and t the time of the measurement. Solving Eq. 2 it results:

$$\widetilde{a}_i(t) = I_\gamma^i A(T_{irr2}) e^{-\lambda_i T} \frac{\left(1 - e^{-\lambda_i t}\right)}{\lambda_i t} \tag{3}$$

Due to the presence of the integral term in Eq. 3 errors are not independent and decrease as a function of time. But this correlation can be calculated and included in the χ^2 minimization procedure of the data:

$$\chi^2 = \sum_{k,l} D_k V_{kl}^{-1} D_l \tag{4}$$

where

$$D_k = a_i(t_k) - \widetilde{a}_i(t_k) \tag{5}$$

and V_{kl} is the covariance matrix between successive times t_k. In a first step, we checked our method on the most dominant γ-ray from ^{51}Ti (E_γ=320.0824 (4) keV, I_γ=93%) which was produced by neutron capture on ^{50}Ti, contained in the titanium holder. We fitted the activity of this γ-ray as a function of time with Eq.3 and extracted a half-life value of (5.82±0.06) min that is in good agreement with the evaluated value (5.76±0.01) min from Xiaolong (2006).

Then we applied our procedure to the γ-ray activities of ^{244}Am shown on Figure 3 as a function of time. The results of the fit procedure are given in Table 1 for each γ-ray. As the isomeric state decays principally to the ground state of ^{244}Cm (71%) and on the first 2+ excited state (27%) emitting a γ-ray of 43 keV (see Figure 1) which is strongly reduced by internal conversion, the intensities of the high energy γ-rays are small and their corresponding counting rate is low as seen on Figure 2. Moreover some of them were interfered with other γ-rays as indicated in Table 1. The maximum interference was observed for the 1041.278 keV γ-ray in which the 1038.760 keV (I_γ =8%) ray from ^{135}I contribute to (5.6±0.5)%. For the others we checked that no peak remains after 10 hours of cooling excluding a lot of contaminants having γ-rays with close energy. In our measurement the 984.91 keV γ-ray which should be associated to the decay of the 984.91 keV level was not observed, confirming the spin of this level to be 0+ as proposed by (Hoff et al. , 1984). For the ground

[1] The uncertainty was certified to be of 1.25% at 3 standard deviations by LEA - CERCA

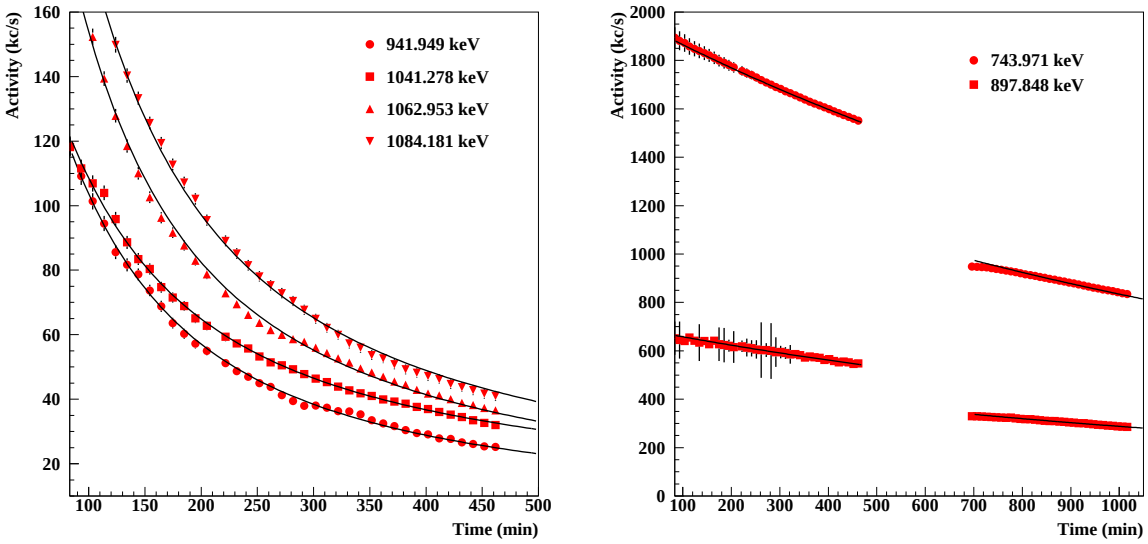

Figure 3: Measured evolution of the γ-ray activities of ^{244m}Am (left) and ^{244gs}Am (right) as a function of time. The activities were corrected from the total efficiency determined with ^{60}Co and ^{152}Eu (see text). The step in the decay of ^{244gs}Am (right figure) is due to the accumulation procedure which creates a remnant activity that disappears when a new run is started. The lines are the results from the fit.

state, the 538.4 keV γ-ray was not clearly visible so that we only estimated a limit on its contribution from the background counting rate.

We finally extracted a mean half-life of (28.3±1.3) min for the ^{244m}Am that is 8.8% higher than the estimated one (26±1) min, and (10.65±0.12) h for the ^{244gs}Am, that is 5.4% higher than the existing one (10.1±0.1) h .

3.2. Capture branching ratio and absolute γ-ray emission probabilities

In order to get an absolute normalization of the γ-ray emission probabilities, we measured separately the total ^{243}Am(n,γ)^{244}Cm and the partial ^{243}Am(n,γ)^{244gs}Am reaction cross sections. We deduced the partial ^{243}Am(n,γ)^{244m}Am cross section from the subtraction of the two previous ones corrected from the 0.0361 (13)% of electron capture forming ^{244}Pu (Akovali , 2003).

	E_γ (keV)	$T_{1/2}$(s)	$a_i(0) = I^i_\gamma A(T_{irr2})e^{-\lambda(t)}$ (kBq)	comment
	941.949 (18)	1825±133	263±18	
^{244m}Am	1041.278 (22)	1915.9±139	236±12	interfered ^{135}I
	1062.953 (18)	1577±79	437±20	
	1084.181 (14)	1700±150	479±37	interfered ^{198}Au
	mean	1695±79		
	538.400 (16)			not visible
^{244gs}Am	743.971 (5)	38578±487	1967±16	
	897.848 (7)	37544±880	695±8	
	mean	38335±426		

Table 1: Results of the minimization procedure with Eq.3 of the γ-activity as a function of time.

The total reaction cross section was measured as in the reference Marie et al. (2006) by counting the number of ^{244}Cm atoms formed in the first irradiation after one day of cooling. At this time, the isomeric state has totaly decreased and 21% of ^{244gs}Am remains, resulting in a correction of 1.77% for the non-complete decay of ^{244}Am onto ^{244}Cm. An example of α-energy spectra is shown on Figure 4 where we see the dominant α-rays of ^{243}Am (E_α= 5275.3 (10) keV, I_α=87.1 (3) %; E_α= 5233.3 (10) keV, I_α=11.2 (3) %) and the α-rays from ^{244}Cm (E_α= 5804.77 (5) keV, I_α=76.90 (10) %; E_α= 5762.64 (3) keV, I_α=23.10 (10) %). A small contamination of ^{242}Cm also appears on the spectra. This contaminant is present in the irradiation channel and settles on the surface of the target explaining the reduced width of the peak distribution as compared to others. The activities were measured by fitting the energy-spectra with the function described in reference Marie et al. (2006).

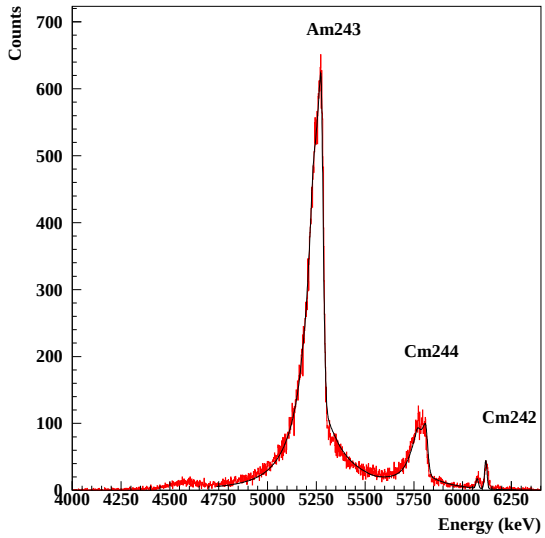

Figure 4: Alpha-energy spectra measured after 21 hours of cooling at a distance of (290±0.01) mm from the target. The result of the fit is also shown.

The neutron fluency seen by the ^{243}Am sample was measured with a Al-1%^{59}Co monitor foil irradiated together with the sample. The effective capture cross section of ^{59}Co was calculated with MCNP code to be (31.1±0.19) b (see Marie et al. 2006) and the deduced fluency from the ^{60}Co γ-activity was of (5.59±0.16) 10^{18} n/cm^2 for the first irradiation and (6.77±0.2) 10^{17} n/cm^2 for the second one. The main errors on the determination of the neutron fluency come from the uncertainty on the number of ^{59}Co contained in the Al foil [2] and from the total efficiency of the HPGe measured with the calibration sources.

As the target burn-up can be neglected, the effective cross section of ^{234}Am can be deduced directly from the ratio of the ^{244}Cm to ^{243}Am activities, corrected from their decay half-life ($T_{1/2}$=7370±15 years for ^{243}Am and $T_{1/2}$=18.11±0.03 years for ^{244}Cm) and using the neutron fluency previously determined. We obtained an effective capture cross section of $\hat{\sigma}_{tot}$ = (73.6 ± 3.2) b confirming the value of (74.1±3.3) b measured in the previous experiment by Marie et al. (2006) and the extracted thermal cross section value at 0.0253 meV: (81.8±3.6) b. These two measurements have approximately the same systematic errors as they use the same set-up, but errors are mainly dominated by non-systematic errors as the samples were different, irradiations were done in different neutron flux conditions, etc.. We then extracted mean effective and thermal values for

[2] certified to be of 2% by the Institute for Reference Materials and Measurements, Geel, Belgium.

the total neutron capture cross section of ^{243}Am considering these two experiments as independent:

$$\hat{\sigma}_{tot} = (73.8 \pm 2.3)b$$

$$\sigma_{tot} = (81.3 \pm 2.5)b$$

The ^{243}Am(n,γ)^{244gs}Am reaction cross section was extracted from the partial activities measured in the previous section (see Table 1). Indeed, as the ground state decay is associated to only one single β-group (Vandenbosch and Day , 1962) populating the 1042 keV level in ^{244}Cm (see Figure 1) which decays to the 4+ ($E_\gamma = 897.848$ keV), 6+ ($E_\gamma = 743.971$ keV) and 8+ ($E_\gamma = 538.4$ keV) members of the ground state rotational band of ^{244}Cm, the partial activities give the relative contribution of each γ-ray to the total β-decay of the ground state. In our data the 6+ $\rightarrow$ 8+ transition was not clearly observed due to the high Compton background, but we have estimated its intensity to be less than 1%. Then using the internal conversion coefficients from (Akovali , 2003) we calculated new absolute intensities per 100 decays (see Table 2). These values are compatible within the error bars with the previous one, increasing slightly the 6+ $\rightarrow$ 6+ transition intensity. We used these new intensities to calculate the ^{243}Am(n,γ)^{244gs}Am cross section from the partial activities by expressing the total activity $A(T_{irr2})$ after the second irradiation:

$$A_{gs}(T_{irr2}) = \lambda_{gs} N(0) \hat{\sigma}_{gs} \phi_2 (1 - e^{-\lambda_{gs} T_{irr2}}) + A_{gs}(T_0) e^{-\lambda_{gs} T_{irr2}} \quad (6)$$

where $N(0)$ is the initial atom quantity of ^{243}Am, $\hat{\sigma}_{gs}$ the effective capture cross section and $A_{gs}(T_0)$ the ^{244gs}Am activity at the beginning of the second irradiation that can be expressed as:

$$A_{gs}(T_0) = \lambda_{gs} N(0) \hat{\sigma}_{gs} \phi_1 (1 - e^{-\lambda_{gs} T_{irr1}}) e^{-\lambda_{gs}(T_0 - T_{irr1})} \quad (7)$$

We see that the effective cross section $\hat{\sigma}_{gs}$ can be factorized in Eq.6 resulting in a proportionality between the activity and the cross section, the proportionality coefficient being only dependent on the time structure of the irradiation-cooling phases, the neutron fluxes and N(0). We found an effective cross section of

$$\hat{\sigma}_{gs} = (3.49 \pm 0.1)b$$

In the previous experiment (Marie et al. , 2006) we published an effective cross section value of (4.7±1.4) b, using the evaluated γ-ray intensities from Akovali (2003) reported in Table 2. Correcting this value from the new intensities and half-life values we reevaluated the cross section to be (3.42±0.1) b that is compatible with the cross section obtained in this work. The error was also reevaluated and reduced, the dominant uncertainties being the errors on the γ-intensities.

From the two previous cross sections we extracted the capture branching ratio to the isomeric state value :

$$BR = 1 - \frac{\sigma^{gs}}{\sigma^{gs+m}} = 0.953 \pm 0.02$$

The errors on the mass of the sample and the neutron flux canceled so that the error is essentially dominated by the determination of the ^{244}Cm activity. Finally we obtained the partial effective and thermal ^{243}Am(n,γ)^{244m}Am cross section values following the same assumptions on the neutron flux than in reference Marie et al. (2006):

$$\hat{\sigma}_m = (70.11 \pm 3.2)b$$

$$\sigma_m = (77.57 \pm 3.5)b$$

This value is little smaller than the recent value obtained by Hatsukawa et al. (1997) (84.44 b) but in the latter reference no uncertainty was published.

Using this effective cross section, we recalculated the new absolute emission probabilities of the γ-rays associated to the isomer (see Table 2). Results are compatible with the previous ones except for the 941.949 keV which reduces its intensity by a factor two. Finally we reduce largely the error bars associated to the absolute intensities.

	$\hat{\sigma}$(b)	E_γ (keV) (Akovali , 2003)	I_γ (Akovali , 2003)	α	I_γ (this work)
^{244m}Am	70.11±3.2	941.949 (18)	0.35 % (13)	0.0157	0.16±0.011%
		1041.278 (22)	0.19 % (8)	0.18 (7)	0.127±0.001%
		1062.953 (18)	0.27 % (9)	0.12 (4)	0.28±0.013%
		1084.181 (14)	0.35 % (13)	0.038 (10)	0.30±0.016%
^{244gs}Am	3.49±0.1	538.400 (16)	0.66 % (22)	0.05	<1.11±0.6%
		743.971 (5)	66 % (24)	0.082 (5)	68.6±0.9%
		897.848 (7)	28 % (11)	0.0172 (4)	24.2±0.3%

Table 2: Measured absolute γ-intensities per 100 decays, energies. Also are indicated the measured conversion coefficient α from Akovali (2003) and the measured effective capture cross sections.

4. Conclusion

The ^{243}Am isotope is an important contributor in high burn-up fuel and/or transmutation systems which is responsible for the formation of ^{244}Cm via the ^{244}Am isotope. Using a high counting rate acquisition system we measured precisely the β-decay half-lives of the ground and isomeric states of ^{244}Am. From the γ- and α-spectroscopy analysis of the irradiated ^{243}Am target we also measured the capture cross section to ^{244}Cm and the branching ratio to the isomeric state. Using these values we also proposed new absolute γ-ray emissions probabilities. We found that our results are globally in good agreement with previous measurements and existing data but we improve seriously the uncertainties on all these quantities.

Acknowledgments

The authors would like to thank the personnel of ILL reactor division for their technical help and support. The authors are also thankful to Dr. David K. Shuh from Lawrence Berkeley National Laboratory for his kind assistance.

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