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PRELIMINARY RESULTS ON ^{241}Am TRANSMUTATION IN A HIGH THERMAL FLUX*

T. Bolognese[a1], G. Fioni[a], M. Cribier[a], F. Marie[a], M. Aubert[c], S. Ayrault[b], J.-M. Cavedon[c], F. Chartier[c], F. Doneddu[c], H. Faust[d], A. Gaudry[b], F. Gunsing[a], Ph. Leconte[d], F. Lelievre[a], J. Martino[a], R. Oliver[d], A. Pluquet[a], M. Spiro[a], C. Veyssiere[a]
[a] *DSM / DAPNIA, CEA/Saclay, 91191 Gif-sur-Yvette, France*
[b] *DSM / DRECAM - CNRS, CEA/Saclay, 91191 Gif-sur-Yvette, France*
[c] *DCC / DPE, CEA/Saclay, 91191 Gif-sur-Yvette, France*
[d] *Institut Laue-Langevin, Grenoble, France*

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The possibility to transmute ^{241}Am by a high intensity thermal neutron flux depends essentially on the value of the ^{242gs}Am capture cross section. As the recommended values given in the two most widely used nuclear data libraries ENDF-B/VI and JEF-2.2 differ by more than a factor of 20, an experiment has been carried out at the Institut Laue-Langevin in Grenoble (France) to fix this discrepancy. The obtained results and their impact on the present understanding of nuclear waste incineration systems will be discussed.

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1 Introduction

The transmutation of radioactive nuclear waste issued from conventional nuclear power plants, constitutes a considerable challenge for the scientific community.

The INCA (Incineration by Accelerator) project [1] of the Directorate for Science of Matter of the French Atomic Energy Authority (CEA/DSM), aims to outline the ideal physical conditions to transmute minor actinides (mainly $^{241-243}$Am, ^{237}Np and $^{244-245}$Cm) and some long living fission products (^{99}Tc, ^{129}I and ^{135}Cs) in an intense neutron flux obtained by an accelerator driven spallation facility coupled to a sub-critical system.

In this framework, a sizeable discrepancy of a factor of 20 was pointed out on the ^{242gs}Am thermal neutron capture cross section from JEF-2.2 [2] (5511 barns). and ENDF-B/VI [3] (252 barns) libraries. For the ^{242gs}Am data, JEF-2.2 is based on the JENDL-3.2 library [4] which rely

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[1] E-mail address: bolognese@dapnia.cea.fr

on a Japanese evaluation [5]. No experimental data are actually available for this reaction and the recommended values are based on theoretical evaluations.

This great discrepancy has no major impact on the present commercial nuclear reactors, where the ^{242gs}Am is present at very low concentration. This is due to the absence of ^{241}Am in the starting fuel[2], to the low neutron flux and to the very short half-life of the ^{242gs}Am (16.02 h). On the other hand, in a minor actinide incineration system the initial fuel will contain mostly ^{241}Am and the high neutron flux will contribute to form a large amount of ^{242gs}Am. The quantitative impact of its capture cross section on the behaviour of an incineration system based on thermal neutrons, can be obtained by looking at the number of neutrons required to incinerate by nuclear fission an atom of a specific nuclear fuel. This parameter, also known as the D-factor, is plotted in Fig. 1 as a function of the thermal neutron flux [6]. A mixture of minor actinides composed by 43% ^{241}Am, 27% ^{243}Am, 17% ^{237}Np and 10% ^{244}Cm, was assumed in the calculations. This composition corresponds to the minor actinides which are supposed to be present after multi-recycling of a MOX (Mixed Oxide) fuel. The continuous and the dashed lines in Fig. 1 are obtained respectively by using the JEF-2.2 and the ENDF-B/VI libraries. A negative value of the D-factor corresponds to a gain of neutrons during the transmutation process and it should be obtained to have a transmutation system economically acceptable. In reality the "break-point", where the system becomes "critical", is situated at about -0.3 to take into account the neutron leakage from the system. From Fig.1, it is therefore evident that the transmutation of a mixture of minor actinides issued from multi-recycling. of MOX fuels, is possible with a thermal neutron flux of about 2×10^{15} n/s/cm^2 only if the value of the ^{242gs}Am capture cross section is lower than a few hundred barns.

A correct determination of this cross section is indeed crucial for the present understanding of minor actinides incineration systems.

2 Experimental procedure

In order to assess experimentally the transmutation efficiency of ^{241}Am in a high thermal neutron flux and to estimate the ^{242}Am capture cross sections, a total of 12 samples containing each about 30 μg of ^{241}Am oxide canned in quartz ampoules have been irradiated at the High Flux Reactor of the Institut Laue-Langevin in Grenoble (France). This single highly enriched fuel element reactor is cooled and moderated by D_2O and provides the highest purely thermal neutron flux available in the world. The combination of a very high neutron flux and the absence of epithermal neutrons, has been essential for the success of the following experiment. The experimental method consists in the measurement of the isotopic evolution of several samples irradiated for different times and in different neutron fluxes. The H9 beam position, which provides the highest neutron flux easily available at ILL, was used for 8 of the 12 samples. The sample changer of the Lohengrin spectrometer was used to bring the samples at a distance of about 60 cm from the fuel element, where the neutron flux approaches 6×10^{14} n/s/cm^2 with a thermalization coefficient of more than 98%. The T4 irradiation position located on the top of the ILL-reactor and giving access to a neutron flux of about 2×10^{13} n/s/cm^2 , was used for four samples. In Table 1 the experimental programme is given with the precise mass of the sample and the irradiation duration.

The main reason for using two different fluxes resides on having two independent sets of data to be used in the final fit and especially to verify that the epithermal component in H9 could be

[2] ^{241}Am is mainly formed by β-decay of ^{241}Pu during the fuel burn-up.

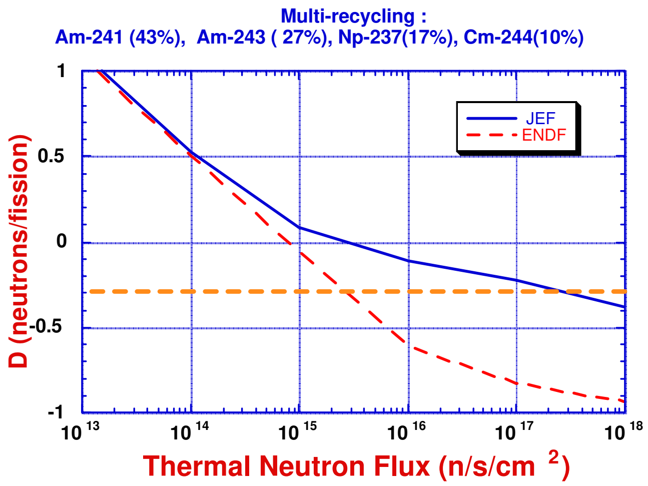

Fig. 1. Number of neutrons required to incinerate by nuclear fission . one atom of the given isotope mixture as a function of the thermal neutron flux intensity. The continuous line is calculated using the JEF-2.2 library, while the dashed line is obtained with ENDF-B/VI. The starting mixture corresponds to the average composition of the minor actinides after multi-recycling of a MOX-fuel.

Table I. Details of the irradiation programme. In three cases, two ^{241}Am samples were irradiated simultaneously to permit two different mass spectrometry analyses, as described in section 3.3. The neutron flux at the H9 position is about 6×10^{14} n/s/cm^2, while on T4 is about 2×10^{13} n/s/cm^2.

^{241}Am Mass [μg]	Irradiation duration [days]	Irradiation Position
29.52	30 minutes	H9
25.94	0.95	H9
30.21	2.01	H9
27.89	5.02	H9
31.52	10.98	H9
25.24	10.98	H9
25.32	19.03	H9
38.14	19.03	H9
37.92	5.01	T4
27.95	11.01	T4
29.81	24.0	T4
29.85	24.0	T4

really neglected in the data analysis. In fact, the T4 position is outside the heavy water reactor vessel at about 1.3 meters from the fuel element, where the neutron flux is perfectly thermalized.

The isotopic chains formed by thermal neutron irradiation of ^{241}Am are shown in Fig. 2. Off-line gamma-ray spectroscopy and mass spectrometry were used to determine the evolution of each irradiated sample as described in section 3.3.

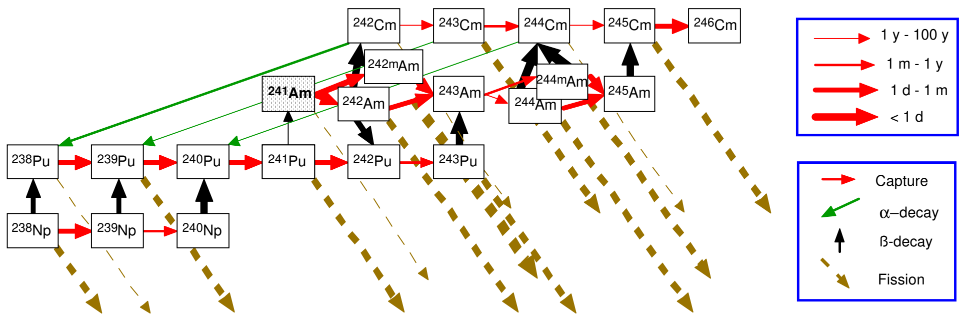

Fig. 2. First steps of the isotopic chain formed during the irradiation of ^{241}Am by thermal neutrons.

3 Off-line measurements

The very short half-life of ^{242gs}Am and its very low concentration, make impossible on-line experiments. Mass spectrometry techniques must then be applied. A number of ancillary measurements are nevertheless necessary to obtain a reasonable precision on the final results.

3.1 Neutron Flux determination

A 20 mg natural iron wire, canned in a quartz ampoule, was irradiated together with each ^{241}Am sample to measure the average thermal neutron flux.

During the irradiation of the iron sample, the isotope ^{59}Fe is formed through neutron capture of ^{58}Fe. ^{59}Fe then decays with a half-life of $T_{1/2}$ = 44.5 days to the stable ^{59}Co through beta decay followed by gamma-ray emission. By measuring the absolute gamma-ray intensity of the 1291.6 keV line of the irradiated iron samples, the average neutron flux has been determined [7] with an accuracy of about 4%, which is satisfactory for the present experiment.

The choice of natural iron as a flux monitor was made in consideration of the experimental conditions. The very high temperature inside the H9 beam tube makes the use of any aluminium alloy impossible and the high neutron flux impose a very ”diluted” sample to avoid to work with a too high radioactivity.

In Fig. 3 the average neutron flux is plotted as a function of the irradiation time, where the 0 value corresponds to the beginning of the irradiation campaign. We observe a steady decrease of the neutron flux as a function of time, probably due to the movement of the reactor control rod during the cycle. The reactor stability is better than 2% over the considered time interval. For the two initial values the reactor was operated at the nominal power, before the decrease at 52 MW from the sixth day on.

3.2 X and γ-ray spectroscopy of irradiated samples

The accuracy of the final results depends strongly on the determination of the capture cross section of ^{241}Am and the corresponding branching ratio towards the ground and the metastable

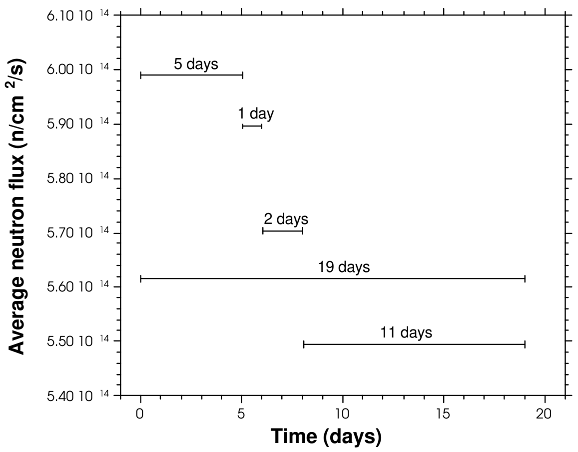

Fig. 3. Average thermal neutron flux at the H9 position of the ILL reactor as a function of the time, where 0 corresponds to the beginning of the irradiation. It should be noted that after 6 days of irradiation, the reactor thermal power was decreased from 57 to 52 MW.

states of ^{242}Am.

To determine these values, dedicated X and gamma-ray spectrometry measurements have been carried out using two samples irradiated respectively 30 minutes and 1 day on H9. The principle relies on the determination of the X and gamma activities after 1, 2 and 273 days from the end of the irradiation. In Fig. 4 a schematic view of the first steps of the ^{241}Am evolution is given. ^{242gs}Am is formed either directly by ^{241}Am(n,γ) or by decay of the ^{242m}Am by internal conversion. The ^{242gs}Am then undergoes β-decay to ^{242}Cm (83%) and electron capture to ^{242}Pu (17%).

The decay rate of ^{242gs}Am can then be determined by looking at the K_α lines of ^{242}Pu at 99.5 and 103.7 keV, and at the 42.1 keV γ-line. Complementary information can be obtained by the α-decay of ^{242}Cm to 238Pu observed by looking at the associated 44.1 and 101.9 keV γ-lines.

At short cooling times, the decay of ^{242gs}Am formed by ^{241}Am(n,γ) dominates, with the contribution coming from ^{242m}Am decay being negligible. X and γ-spectroscopy measurements will then provide directly the capture cross section for ^{241}Am(n,γ)^{242gs}Am.

These measurements require a very short irradiation (30 minutes) in order to avoid a huge background coming from the X-rays emitted from the fission fragments.

At long cooling times, the "original" ^{242gs}Am has disappeared ($T_{1/2}$ = 16.02 h) and the ^{242gs}Am present in the sample is formed only by the Internal Conversion of ^{242m}Am ($T_{1/2}$ = 141 y). In this case, X and γ-spectroscopy measurements of a sample irradiated for 1 day on H9 will provide the capture cross section for ^{241}Am(n,γ)^{242m}Am.

The X and γ spectra after a cooling time of 1.9 and 273 days were obtained. The γ-transition at 42.1 keV is strongly suppressed after 273 days as a consequence of the disappearing of the

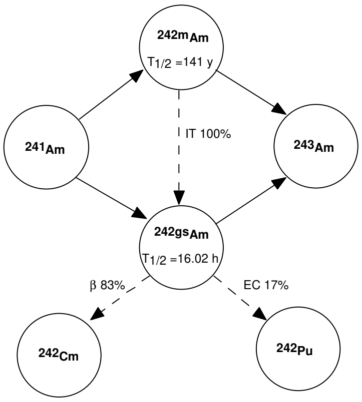

Fig. 4. Schematic view of the first steps of the ^{241}Am transmutation chain.

^{242gs}Am formed during the irradiation.

The final results are :

^{241}Am(n,γ) = (69530) barns

and

$\frac{\sigma_c(^{241}\mathrm{Am}\rightarrow^{242gr}\mathrm{Am})}{\sigma_c^{tot}} = 0.91 \pm 0.01$.

While the branching ratio is in agreement with the literature value, the total capture cross section is 16% higher than the recommended JEND-3.2 valuation and 22% lower than a recent Japanese measurement [9].

3.3 Mass spectrometry analysis

All samples have been analyzed by Thermal Ionization Mass Spectrometry (TIMS). With this technique, the Am isotope ratios for all samples, as well as those of Cm and Pu for some samples, have been measured on some tens of ng of each element, with a precision ranging from 0.5 % to some percents, depending on the value of the isotopic ratios. For some samples, the isotope dilution analysis with tracers has been applied for the determination of Cm and Pu amounts relative to the initial ^{241}Am quantity.

Due to the isobars coming from the different elements, it was necessary to perform a separation between these elements before TIMS analyses. These separations have been carried out by High Performance Liquid Chromatography (HPLC), so that the composition of different Am, Cm and Pu isotopic chains has been determined with very high accuracy and precision.

In Fig. 5, the isotopic ratio ^{243}Am/^{241}Am is plotted as a function of the irradiation time. The continuous and the dotted lines are respectively calculated for a nominal neutron flux of 5×10^{14} n/s/cm^2 from JEF-2.2 and ENDF-B/VI libraries. The uncorrected experimental data are plotted with a square marker connected by a dashed line to guide the eye. It is evident from these raw data that the value of the ^{242gs}Am cross section is of the order of the ENDF-B/VI evaluation.

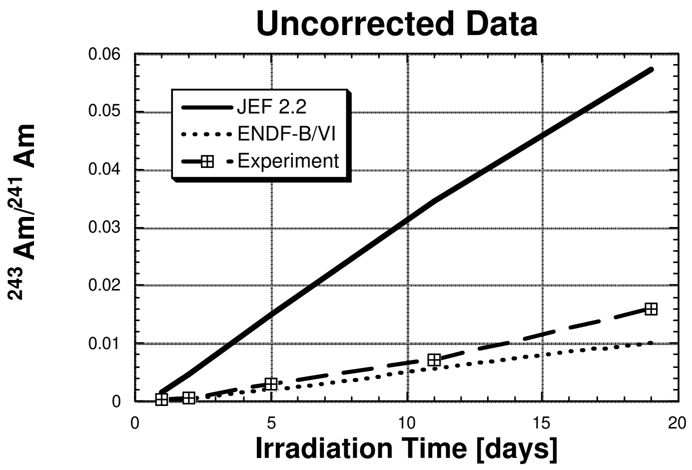

Fig. 5. Isotopic ratio ^{243}Am/^{241}Am plotted as a function of the irradiation time. The continuous and the dotted lines are respectively calculated for a nominal neutron flux of 5×10^{14} n/s/cm^2 from JEF-2.2 and ENDF-B/VI libraries. The uncorrected experimental data are plotted with a square marker connected by a dashed line to guide the eye.

A qualitative and semi-quantitative analysis (15 to 20 % uncertainty) has also been carried out by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) to obtain the fission fragment yields. At present the results are still under evaluation and a comprehensive article will be submitted within few months.

4 Results and conclusions

The mass spectrometry data have been analyzed to determine the capture cross sections of ^{242gs}Am. The measured isotopic ratios (^{243}Am/^{241}Am and ^{242m}Am/^{241}Am) for all samples, as well as the Cm, Pu and fission products contents for two samples, have been compared in a global fit with the prediction obtained by solving the Bateman equations describing the evolution of the samples. The values of the ^{241}Am(n,γ) and the corresponding branching ratio were taken from the γ-ray spectroscopy measurements described in the previous section. The preliminary result is:

$^{242gs}\mathrm{Am}(n,\gamma) = (210 \pm 50)$ barns.

An accuracy of 25% is given as the result is still preliminary and further data analysis is still in progress. The preliminary result is in agreement with both the ENDF-B/VI value and with a recent Belarussian evaluation [10]. The option to transmute ^{241}Am together with the minor actinides issued from multi-recycling of a MOX fuel by an intense flux of thermal neutrons is still theoretically open.

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