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Benoit Geslot[1,*], *Adrien* Gruel[1], *Paul* Ros[1], *Patrick* Blaise[1], *Pierre* Leconte[2], *Gilles* Noguere[2], *Ludovic* Matthieu[3], *David* Villamarin[4], *Vicente* Becares[4], *Arjan* Plompen[5], *Stefan* Kopecky[5], and *Peter* Schillebeeckx[5]

[1]CEA, DEN, DER/SPEx, Cadarache, F-13108 St Paul Lez Durance, France
[2]CEA, DEN, DER/SPRC, Cadarache, F-13108 St Paul Lez Durance, France
[3]CENBG, CNRS/IN2P3-Université de Bordeaux, Chemin du Solarium B.P. 120, 33175 Gradignan, France
[4]CIEMAT – Nuclear Fission Division – Avenida Complutense, 40, 28040, Madrid, Spain
[5]EC-JRC-IRMM, B-2440 Geel, Belgium

Abstract. An experimental program, called AMSTRAMGRAM, was recently conducted in the Minerve low power reactor operated by CEA Cadarache. Its aim was to measure the integral capture cross section of [241]Am in the thermal domain. Motivation of this work is driven by large differences in this actinide thermal point reported by major nuclear data libraries. The AMSTRAMGRAM experiment, that made use of well characterized JRC-IMM americium samples, was based on the oscillation technique commonly implemented in the Minerve reactor. First results are presented and discussed in this article. A preliminary calculation scheme was used to compare measurements to calculated values. It is shown that this work confirms a bias previously observed with JEFF3.1.1 (C/E-1 = -10.5 ± 4 %). On the opposite, the experiment is in close agreement with [241]Am thermal point reported by JEFF3.2 (C/E-1 = -0.5 ± 4 %).

1 Introduction

For nearly 10 years, several experiments have been conducted to improve [241]Am radiative capture cross section. Indeed, evaluated cross sections from major libraries differ from 10 % to 15 %, especially in the thermal domain [1-2]. For instance, a difference of 9 % on the thermal point can be observed between JEFF3.2 and ENDF-B.VII.1 nuclear data libraries. An international collaborative working group organized by NEA-OECD started in 2015 with the aim of explaining such discrepancies [3].

In this framework, an experimental program was recently conducted by CEA Cadarache in the Minerve reactor. Its main objective was to measure [241]Am integral capture cross section in a very thermal neutron spectrum, providing valuable information on the [241]Am thermal point, with a target accuracy of 3 % (1σ).

This experiment made use of 7 americium samples manufactured by ITE Karlsruhe in the framework of a previous collaboration between JRC-IRMM and CEA [4]. Measurements setup was based on the reactivity oscillator technique commonly implemented in Minerve [5]. Gold and lithium samples with the same geometry as americium samples were used for calibration.

2 Experimental setup

2.1 Reactor configuration

Minerve is a pool type reactor operated at low power (100 W maximum). Its core is made of two parts:

- a driver zone (DZ) with highly enriched uranium/aluminum assemblies and surrounded by graphite reflector blocks ;
- an experimental zone (EZ) in which 3% enriched UO_2 fuel rods are loaded in a square lattice with a pitch of 1.26 cm.

At the center of the experimental zone, an irradiation channel makes it possible to introduce various material samples in the core. Samples are held in an oscillator that is used to perform pile oscillator experiments. Samples reactivity worth (in the range of a few pcm) are obtained and compared against reference materials.

In the AMSTRAMGRAM configuration, a water hole is arranged at the center of the experimental zone. Its dimensions (roughly 4.4 cm in radius) were chosen so as to maximize the moderation ratio at the oscillation location.

The neutron spectrum in the oscillation channel exhibits a strong thermal component, as shown on Fig. 1. The reactivity sensitivity to the americium cross section is also plotted (dotted curve). It shows that the thermal

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[*] Corresponding author: benoit.geslot@cea.fr

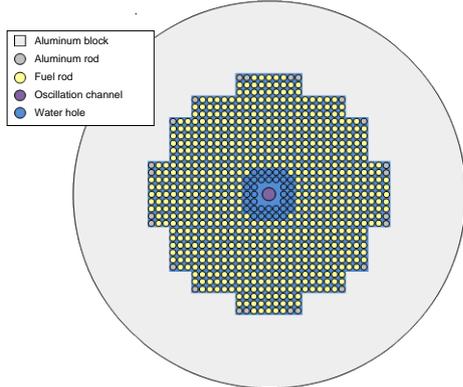domain (below the first resonance at 0.3 eV) accounts for around 80 % of the reactivity.

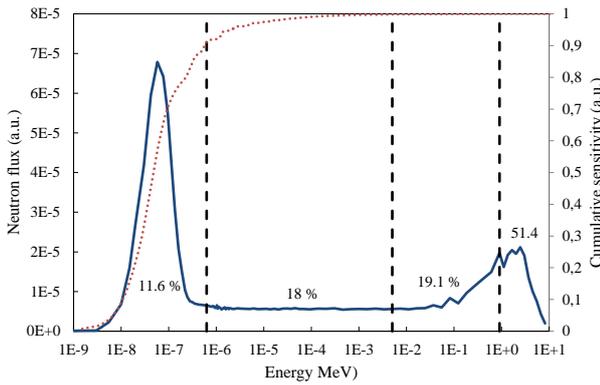**Fig. 1.** Drawing of the AMSTRAMGRAM experimental zone.

Fig. 2. Neutron spectrum in the oscillation channel (solid curve) and cumulative sensitivity of core reactivity to ^{241}Am capture cross section (dotted line).

2.2 Americium samples

Americium samples used in this work were manufactured by ITE Karlsruhe from on raw material given by CEA Marcoule. They were previously irradiated at JRC-IRMM for the measurement of the ^{241}Am(n,2n) cross section [4].

Samples are made of ^{241}AmO$_2$ and Al$_2$O$_3$ powders mixed and pressed into pellets or 12 mm in diameter and 2 mm thickness and then encapsulated in aluminum containers (see drawing on Fig. 3).

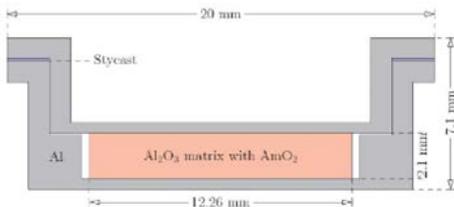Americium mass is around 40 mg diluted in roughly 400 mg of alumina.

Fig. 3. Drawing of JRC-IRMM americium samples

2.3 Additional samples

Calibration of the measurement process is obtained by using disks of reference material in dummy containers. Gold disks (Ø10 mm, 0.125 mm thick) and lithium disks (Ø10 mm, 0.2 mm thick) of high purity for purchased.

Additional samples loaded with 400 mg alumina pellets were also measured in order to obtain the reactivity per gram of alumina matrix.

3 Data processing

3.1 The reactivity oscillator technique

Oscillation techniques are based on creating a weak and localized perturbation in a characterized neutron flux. Its characteristics (amplitude, neutron spectrum) can be connected to physical parameters of the perturbation source (i.e. isotopic cross sections).

Oscillation techniques can be classified in function of 3 main parameters: use of a power compensation system, the oscillator frequency, the detector location. Detailed information can be found in the literature [6].

The technique performed in Minerve is called « reactivity oscillator ». It is based on the compensation of the reactivity perturbation induced by the sample motion in the core. The reactor is kept critical using a rotating cadmium rod, which is driven by a monitor boron chamber, located in the reflector, through a retro-active loop.

The measured signal is given by the cadmium rod rotation angle. It is proportional to the small sample reactivity worth. This can be expressed using the exact perturbation theory (EPT).

Let 1 be a reactor state perturbed by a small reactivity sample and 0 be the reference state. P and L refer to operators of production and absorption, $\lambda = 1/k$ where k is the multiplication factor and Ψ and Ψ^+ are direct and adjoint fluxes. Then, the reactivity difference between the two reactor states can be expressed as:

$$\delta\rho = \frac{\left\langle \Psi_0^+ \mid (\lambda_0 \delta P - \delta L)\Psi_1 \right\rangle}{\left\langle \Psi_0^+ \mid P_1 \cdot \Psi_1 \right\rangle} \tag{1}$$

Making the assumption that the test sample is a pure absorber, a first order approximation gives:

$$\delta\rho \approx \frac{\left\langle \Psi_0^+ \mid -\delta L\Psi_0 \right\rangle}{\left\langle \Psi_0^+ \mid P_0 \cdot \Psi_0 \right\rangle} \tag{2}$$

In the reference state, which is the same for every sample, the irradiation channel is filled with an aluminum rod. It is then possible to calculate ratios of $\delta\rho$ against a reference material, like gold or lithium:

$$\frac{\delta\rho}{\delta\rho_{ref}} = \frac{\iint \Psi_0^+ \sigma(E)\Psi_0 dr dE}{\iint \Psi_0^+ \sigma_{ref}(E)\Psi_0 dr dE} \tag{3}$$

Introducing a correction factor $f(\sigma)$:

$$f(\sigma) = \frac{\iint \Psi_0^+ \sigma(E)\Psi_0 dr dE}{\iint \sigma(E)\Psi_0 dr dE}, \tag{4}$$

equation (3) can be rewritten as follows:

$$\frac{\delta\rho}{\delta\rho_{ref}} = \frac{f(\sigma)}{f(\sigma_{ref})}\frac{\iint\sigma(E)\Psi_0 drdE}{\iint\sigma_{ref}(E)\Psi_0 drdE} \qquad (5)$$

In the previous equation, the left hand side term is directly obtained from the experiment. The right hand side term is divided into two factors. The first one can be calculated using a deterministic code like APOLLO2 in a 2D geometry and the second one can be precisely calculated from a TRIPOLI4.10 simulation.

3.2 Measurement setup
One americium sample is worth about -0.2 pcm in reactivity, which is quite low compared to the measurement range (~10 pcm).

In order to increase the signal to noise ratio of the measurement, several samples were oscillated at the same time. For that purpose, up to 5 americium samples (and up 7 gold samples) were loaded column-wise in a sample container. The container was made of aluminum alloy 5754 and sealed by screw-in plugs. It was placed in an oscillator tube along with aluminum rods (alu 5754 as well) so that its mid-plane was aligned with the core mid-plane when the oscillator was in its lower position.

Measurements were composed of sets of 5 oscillation cycles with 120 s period. Acquisition was trigged by the oscillator control system and recorded data included 3 analog signals: oscillator position, current of the boron chamber and angle of the cadmium rod versus time.

Every measurement gave 5 values of the rod angle difference between upper and lower position of the sample in the core. For each oscillator loading, measurements were reproduced at least 5 times. If results were consistent (based on a χ^2 test), measurements were averaged to obtain a mean value.

To extract the reactivity worth of an isotope of interest, it is necessary to subtract any signal coming from the matrix (alumina) and the container (clad and aluminum wedges). Indeed, at first order, the overall reactivity can be linearly divided into a sum of individual components:

$$\delta\rho_{sample} = \delta\rho_{Am} + \delta\rho_{alu} + \delta\rho_{clad} \qquad (3)$$

Last terms in eq. (3) can be obtained by oscillating dedicated samples (empty clads or samples loaded with alumina only). Corrections should take into account any significant mass differences between samples.

3.2 Uncertainties management
Sources of uncertainties that affect the measurement are numerous and difficult to model. They come from mechanical fluctuation in the oscillator, electronic perturbation in signal recording, core evolution during the program, etc.

An empirical approach is used to estimate two overall sources of uncertainties: one responsible for variation amongst oscillation cycles, referred to as "repeatability", and another one responsible for any fluctuation from one measurement to the other, called "reproducibility". Those uncertainty sources are supposed to be independent from signal level. Measurements are also assumed to be uncorrelated.

Under those assumptions, the repeatability can be estimated as the intra-class variance of oscillation cycles $S_{ij}(\varepsilon)$ and the reproducibility can be viewed as the inter-class variance of the whole measurements $S_i(\varepsilon)$.

$$\hat{\sigma}_e^2 = \frac{1}{p\cdot N-1}\sum_{i,j,\varepsilon}\left(S_{ij}(\varepsilon)-\overline{S}_i(\varepsilon)\right)^2 \qquad (6)$$

$$\hat{\sigma}_c^2 = \frac{1}{N-1}\sum_{\varepsilon}\sum_i\left(\overline{S}_i(\varepsilon)-\overline{S}(\varepsilon)\right)^2 \qquad (7)$$

$$\overline{S}(\varepsilon) = \frac{1}{n_\varepsilon p}\sum_{i,j}S_{ij}(\varepsilon) = \frac{1}{n_\varepsilon}\times\sum_i\overline{S}_i(\varepsilon) \qquad (8)$$

The final uncertainty affecting the reactivity worth of sample ε from n measurements of p cycles is expressed as:

$$\hat{\sigma}_\varepsilon = \sqrt{\frac{\hat{\sigma}_e^2}{n\cdot p}+\frac{\hat{\sigma}_c^2}{n}} \qquad (9)$$

3.3 Experimental results
Raw signals (in angular unit) are converted in reactivity unit (cent) using a calibration coefficient. It is obtained by measuring the core reactivity (by reactor doubling time) at various cadmium rod angular positions. This coefficient may be somehow imprecise but note that is has no impact on reactivity ratios.

Results are given in the Table 1. The value given for [241]Am is obtained by subtracting the reactivity from the alumina matrix, which amplitude is about 2 % (positive) of the overall signal (negative).

Table 1. Reactivity per mass unit for test materials.

Material	Mass (mg)	Mass reactivity (cent/g)
Al_2O_3	396	0.009
[197]Au	185.4	-0.707
[nat]Li	83.8	-12.54
[241]Am	41.5	-4.442

When several samples were piled in a sample container, a small shadow effect between samples was observed in the case of lithium and gold. This effect was corrected by extrapolating the reactivity per mass unit measured for different sample loadings down to the average sample mass.

Final mass reactivity ratios is 6.27 in the case of americium / gold and 2.85 in the case of americium / lithium.

4 Preliminary feedback on nuclear data

4.1 Calculation scheme

A two-step preliminary calculation scheme was built to calculate the right hand side terms of equation (5).

In a first step, deterministic code APOLLO2 is used to calculate direct and adjoint fluxes and reaction rate of samples in a simplified 2D reactor model of the experimental zone. Factors defined in equation (4) are obtained for each oscillated sample.

In a second step, a simplified calculation based on TRIPOLI4.10 Monte Carlo code is used to calculate reaction rates in 3D realistic sample models. A neutron spectrum previously obtained from a 3D core Monte Carlo calculation is used as an input of that simplified calculation.

4.2 Preliminary calculated values

In a more evolved interpretation scheme, several correction factors should be applied in order to get rid of modelling biases and obtain more precise results.

A clear trend can still be derived from this preliminary scheme. Indeed a 10 % difference is obtained in the C/E-1 value when comparing calculation using libraries JEFF3.1.1 and JEFF3.2. This confirms a similar trend observed in UOx-PWR and MOx-PWR spectra obtained from the OSMOSE program performed in Minerve [7-8].

Moreover, it is observed that the preliminary JEFF3.2 comparison with experiment is very close to 0. Since reactivity sensitivity to ^{241}Am capture cross section is around 0.8 for the thermal domain, this result support very closely ^{241}Am thermal point as reported by JEFF3.2.

Table 2. Calculation over experiment for JEFF3.1.1 and JEFF3.2.

Library	Thermal point (b)	Resonance Integral (b)	C/E – 1 (%)
JEFF3.1.1	662.549	1689,17	-10.5 ± 4
JEFF3.2	764.397	1995,96	-0.5 ± 4

5 Conclusions

This paper presents preliminary experimental results from the AMSTRAMGRAM program conducted by CEA in the Minerve reactor. A very thermal neutron spectrum was designed for the experiment that aimed at providing a new integral measurement of ^{241}Am radiative capture cross section with target uncertainty of less than 3 %. The experiment, based on the pile oscillation

technique, was only possible thanks to JRC-IRMM that provided several well characterized americium samples.

Analysis shows that a clear trend when comparing measurements to calculations associated to JEFF3.1.1 and JEFF3.2 libraries. First results are in very good agreement with JEFF 3.2 thermal point (662.5 b).

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