Neutron activation of natural materials in a PWR spectrum feedback on ^{116m}In relative γ emission intensities and half-life

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***Abstract*— During the MAESTRO program, carried out between 2011 and 2014 in MINERVE zero power reactor, common Gen-II and Gen-III light water reactor materials were irradiated. For some of these materials, the decay of their activation products was also measured by γ spectrometry. Initially devoted to the measurement of the integral capture cross section by activation and reactivity-oscillation method, these results can also provide useful information on decay data of various radionuclides. This led to a common roadmap shared by the Experimental Physics Section and the Henri Becquerel National Laboratory to improve decay data in nuclear data libraries.**

Results discussed in this paper concern the relative emission intensities of the main γ rays of ^{116m}In. Six irradiations of samples with various physical forms of ^{nat}In were carried out. Measurements were analyzed using decay data from several evaluations and it is shown that γ ray activities are not consistent. Analyses were carried out to provide new relative γ emission intensities from these measurements. The ^{116m}In half-life has also been measured and shows a good agreement with existing values (< 0.1% discrepancy).

Finally, an overview of the foreseen results on additional decay data from the MAESTRO program is given.

***Index Terms*—Decay data, Gamma emission intensities, Half-life, Indium-116m, MINERVE**

I. Introduction

During the MAESTRO program [1], carried out between 2011 and 2014 in MINERVE zero power reactor [2], common Gen-II and Gen-III light water reactor (LWR) materials were irradiated. For some of these materials, the decay of their activation products was also measured by γ spectrometry. Initially devoted to the measurement of the integral capture cross section by activation and reactivity-oscillation methods, these results can also provide useful information on decay data of various radionuclides (RNs).

In this study are presented preliminary results on the relative γ emission intensities and the half-life of ^{116m}In, produced by neutron radiative capture on ^{115}In.

The first part of this paper describes the MINERVE reactor as well as the MAESTRO program. The measurement set up and the data analyses are also presented. The second part focuses on the results obtained for ^{116m}In. The conclusion mentions the foreseen results on additional RNs from the MAESTRO program.

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II. Experimental Setup

A. The MINERVE reactor and the MAESTRO Program

MINERVE is a pool-type Zero Power Reactor (maximum power 100 W) located at CEA Cadarache. It is a coupled thermal-thermal core. The driver zone is loaded with highly enriched uranium fuel plates, and the central experimental zone loaded with about 800 standard 3%-enriched UO_2 fuel pins. At the center of the experimental zone, a vertical channel enables the passing of a device called the oscillation tube, inside which is inserted the sample to be irradiated at the mid-core plane. In this configuration, the neutron spectrum at the irradiation position is close to the one of a standard LWR. About 15.5% of the neutrons are below 0.63 eV, 39% are in the 0.63 eV to 100 keV range, and 45.5% are above 100 keV.

The MAESTRO program aimed at measuring integral (n,γ) cross sections of widely used materials in LWRs. About 40 samples of structural materials (Sn, Al, Fe, …), industrial alloys (Al5754, stainless steel, Zy4, M5), moderators (C, H_2O, CH_2,…) and neutron absorbers (Dy, Er, Gd, …) were irradiated. When applicable, some of these samples were afterwards measured by γ spectrometry.

To test the possibility of using these measurements as sources for new decay data evaluations, analysis were carried out on ^{116m}In. Six samples of three different physical forms were irradiated:

- powder mixed with an alumina (Al_2O_3) powder matrix, inside an aluminum cladding (10-cm long and 1-cm diameter) , labelled “M-In”;
- liquid acid solution (H_2O + 4% HNO_3), with Zy4 and aluminum cladding (10-cm long and 1-cm diameter), labelled “M-In-2”;
- four thin metallic foils (0.25-mm thick, 8-mm in diameter), two with and two without a cadmium shield, labelled “Dosi-In-Cd” and “Dosi-In” respectively.

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B. Measurements and Data Analysis

After irradiation, RN decay is measured on a calibrated high-purity germanium planar 40 cm^3 crystal, with a resolution about 1 keV at low energies. It has been calibrated using standard gamma sources (^{241}Am, ^{109}Cd, ^{57}Co, ^{139}Ce, ^{51}Cr, ^{85}Sr, ^{137}Cs, ^{54}M, ^{88}Y, ^{65}Zn and ^{60}Co) on the [50–1850] keV energy range. The detector is shielded with a 4-cm thick tungsten block, to minimize the background noise. A lead collimator ensures measurement of the central part of the cylindrical samples only. Samples are held above the detector in a rotary device, to take into account potential heterogeneity such as radial activity gradients in the material (Fig. 1).

The measurement time and the number of acquisitions are adjusted based on the RN half-life and activity in order to minimize statistical uncertainties. For ^{116m}In, Total acquisition duration was about ten half-lives (54.2 min), split in 1800 s or 3600 s measurements. The first acquisition started less than 1 hour after the end of irradiation. Typical γ ray spectra measured about 3 hours after the end of irradiation are shown in Fig. 2.

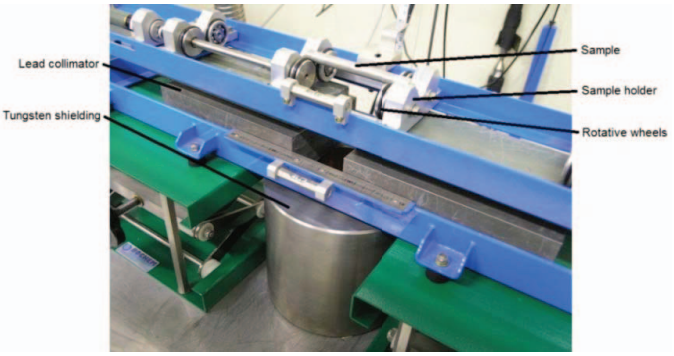

Fig. 1. Picture of the γ spectrometry bench with a 10-cm long sample.

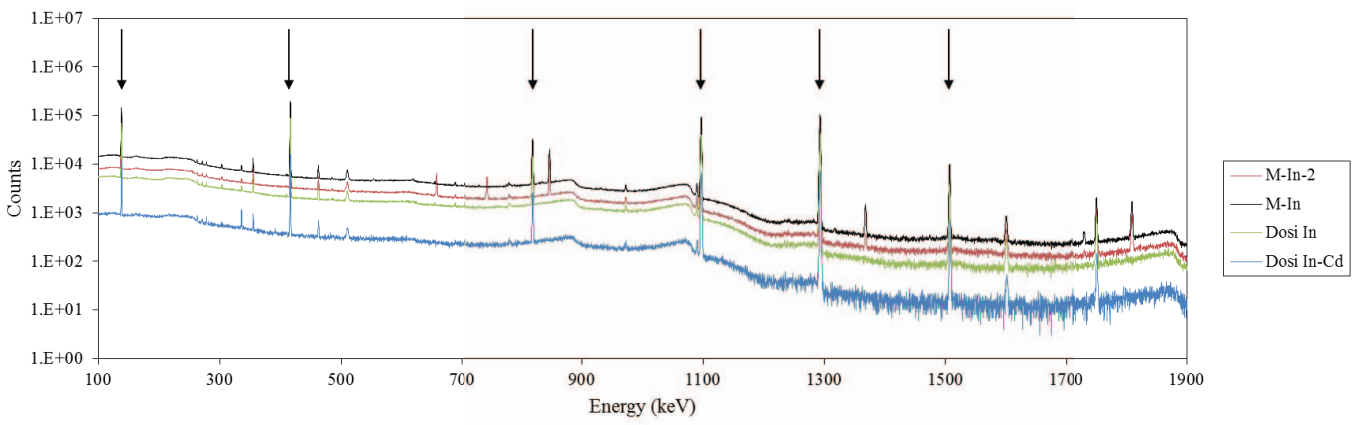

Fig. 2. γ ray spectra on four of the samples, about 3 h after irradiation (1800 s measurements, except for M-In-2: 3600 s).

Spectra were analyzed with the Genie^{TM}2000 software, developed by Canberra [3]. The six most intense γ rays of ^{116m}In in the calibration range are listed in Table I.

TABLE I
INTENSITIES, PER 100 DECAYS, OF THE MOST IMPORTANT ^{116M}In γ RAYS [5] (UNCERTAINTIES AT 1 σ)

Energy (keV)	I_γ (%)
138.326(8)	3.29(13)
416.86(3)	27.7(12)
818.7(2)	11.5(4)
1097.3(2)	56.2(11)
1293.54(4)	84.4(17)
1507.4(2)	9.96(34)

III. Results

A. γ Emission Intensities

The activity measured for a given γ ray at the end of irradiation is given by:

$$A_\gamma = \frac{n_\gamma}{I_\gamma R_\gamma T_\gamma} C_{dec} C_\theta = \frac{N_\gamma}{I_\gamma R_\gamma T_\gamma} \quad (1)$$

with n_γ the net peak area, N_γ the-end-of irradiation count rate, R_γ the detection efficiency, T_γ the self-attenuation of the γ ray in the sample, C_{dec} the decay factor and C_θ the dead-time correction factor. Since the activity is the same for all γ rays, the relative γ emission intensities can be expressed as:

$$\frac{I_\gamma}{I_{\gamma,0}} = \frac{N_\gamma}{N_{\gamma,0}} \frac{R_{\gamma,0}}{R_\gamma} \frac{T_{\gamma,0}}{T_\gamma} \quad (2)$$

where the subscript 0 refers to the reference γ ray. In this preliminary study on the relative γ emission intensities, only

the four metallic foil measurements were analyzed.

Emission intensities of the γ rays are measured relatively to the 1294 keV one because it has the highest γ emission intensity and is supposed to be the best known. Overall uncertainties (1 σ) on this ratio include contributions from:

- end-of-irradiation count rate (between 0.1% and 0.3%);
- detection efficiency ratios (between 0.1% and 1%);
- Monte Carlo γ self-attenuation calculation (between 0.2% and 0.5%).

The end-of-irradiation count rate uncertainty takes into account the counting statistics, the fitting procedure of the Génie^{TM}2000 software if required, the uncertainty on the half-life and the dead-time correction factor (0.5%). For each γ ray end-of-irradiation count rate measurement, about ten successive measurements are combined, leading to a final reduced uncertainty between 0.1% and 0.3%.

For every γ ray, measurements between foils show dispersion below 0.5%.

These results are compared to two evaluations and a measurement from G. Wurdiyanto *et al.* [4]. The first evaluation is taken from Nuclear Data Sheets [5], and the second one from the LNHB [6].

Results are very consistent with data from [4] and [5] except for the 818.7 keV γ ray: (−3.0 ± 1.4)% and (−3.5 ± 0.8)% discrepancy respectively. Another noticeable discrepancy appears for the 138.3 keV γ ray against data from [6]: (−9.4 ± 4.5)%. All other values are consistent within two standard deviations (Table II and Fig. 3).

Nevertheless, further comprehensive uncertainty propagation should be done, taking into account correlations between measurements. It would also be of interest to include to this study the measurements on the two cylindrical samples "M-In" and "M-In-2".

TABLE II
RELATIVE γ EMISSION INTENSITIES (UNCERTAINTIES AT 1 σ)

Energy (keV)	This work	Wurdiyanto [4]	NDS [5]	LNHB [6]
138.326	4.30(2)	4.31(4)	4.36(10)	3.90(17)
416.86	31.91(17)	31.65(23)	32.1(3)	32.8(16)
818.7	14.81(6)	14.37(19)	14.3(1)	13.63(55)
1097.3	69.50(30)	68.73(42)	69.0(7)	66.6(19)
1293.54	100.0(3)	100.00(45)	100(1)	100.0(20)
1507.4	11.780(40)	11.610(88)	11.7(1)	11.80(47)

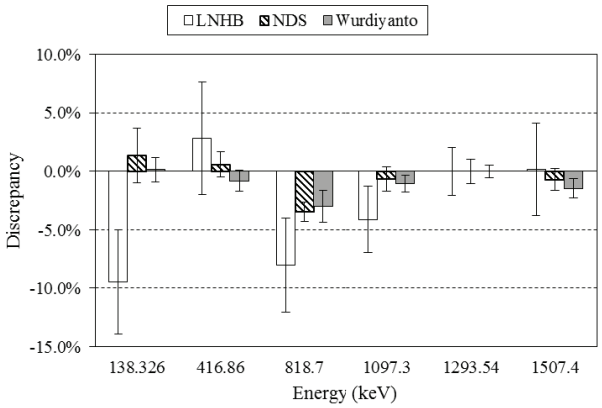

Fig. 3. Discrepancies of the relative gamma probabilities from this work against existing evaluations (uncertainties at 1 σ).

B. *Half-Life*

For half-life determination, all data were firstly analyzed by a fit of the count rate versus time (exponential decay). For some measurements, this led to inconsistent normalized residuals, and more specifically for early measurements (Fig. 4). To overcome this issue, we have modified the fit function: since the count rate is an average over acquisition duration, the fit function chosen in this study is the following:

$$\frac{1}{t_{i+1}-t_i}\int_{t_i}^{t_{i+1}} e^{-\lambda t}\,\mathrm{d}t = -A\frac{e^{-\lambda t_{i+1}}-e^{-\lambda t_i}}{\lambda(t_{i+1}-t_i)} \qquad (3)$$

where t_i is the starting time of acquisition i. The two fitted parameters are the amplitude A and the decay constant λ. Using this method rather than a fit on the count rate, normalized residuals are significantly reduced for some of the measurements (Fig. 5). For consistency, all data were treated the same way, using Eq. (3).

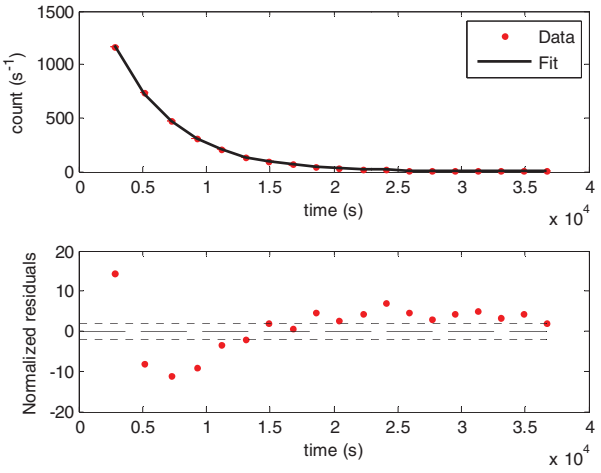

Fig 4. Example of fitted count rate and normalized residuals (417 keV γ ray of the liquid sample M-In-2).

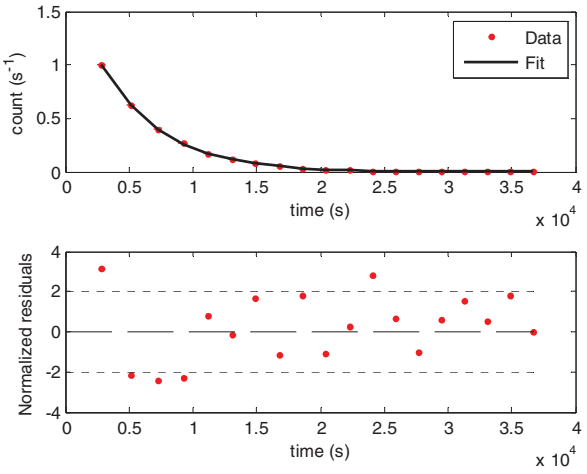

Fig. 5. Example of fit using Eq. (3) and normalized residuals (417 keV γ ray of the liquid sample M-In-2).

Data are fitted using a weighted least-square fit method. Uncertainty on each measurement data is given by the γ spectrometry analysis software. It takes into account the background subtraction, the continuum region on the sides of the peak and the shape of the background. In this preliminary work, uncertainties were considered all uncorrelated. Therefore, the variance on the calculated parameters is:

$$\mathbb{V} = (\mathbb{J}^{T}\mathbb{W}\mathbb{J})^{-1}, \text{ with } \mathbb{W} = \begin{bmatrix} \frac{1}{\sigma_1^2} & \cdots & 0 \\ \vdots & \ddots & \vdots \\ 0 & \cdots & \frac{1}{\sigma_N^2} \end{bmatrix} \qquad (4)$$

where $\mathbb{J}$ is the Jacobian matrix of the system and $\mathbb{W}$ the matrix of the weights of the N data points.

The $T_{1/2}$ values obtained are shown in Fig. 6. The average value of the radioactive half-life of ^{116m}In is 54.24(8) min (weighted mean of all values). Given that the propagated uncertainty considering independent measurements seems very low (0.01%), it has been chosen to keep the standard deviation between all values as the final uncertainty (0.15%). This value is consistent with the recommended value of 54.29(17) min [6], and a recent measured value of 54.20(7) min [7].

This analysis should be continued, especially concerning the uncertainty budget, since all uncertainty components were considered uncorrelated, while all measurements were carried out on a same detector.

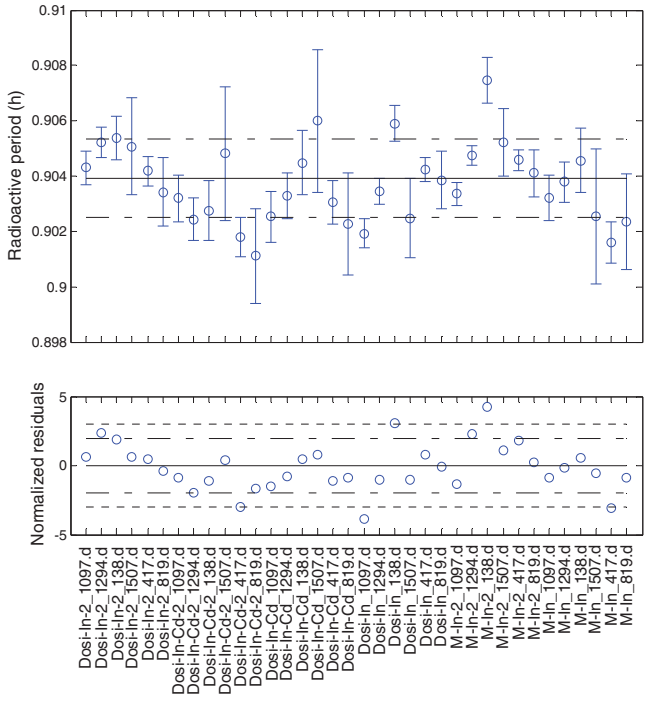

Fig. 6. Measured radioactive half-lives for each γ ray of each sample (top) and normalized residuals (bottom). Top figure: in dashed-dotted-line the standard deviation between all values.

IV. Conclusion & Outlooks

Amongst 40 samples irradiated in the MINERVE reactor during the MAESTRO program, nine samples were measured by γ spectrometry, leading to the characterization of several RNs: ^{152gs,m}Eu, ^{154}Eu, ^{101}Mo, ^{99}Mo, ^{113}Sn, ^{117m}Sn, ^{123m}Sn, ^{52}V, ^{65}Zn, ^{69m}Zn, ^{95}Zr, ^{97}Zr, ^{114m}In, ^{116m}In, ^{134gs,m}Cs, ^{108}Ag and ^{110m}Ag. In order to test the possibility of using these measurements as inputs for new decay data evaluations, a test analysis was carried out on ^{116m}In.

Relative γ emission intensities show a good consistency with existing evaluations. Further work on those data should include the cylindrical liquid and powder samples. In addition, radioactive half-life measurements of this nuclide led to a preliminary value of 54.24(8) min, consistent with most recent values in the literature.

On both studied parameters, a more comprehensive uncertainty study including the sample technological uncertainties and sensitivity analyses should also be led.

However, this work clearly shows that capture rate measurements carried out during the MAESTRO program can be used for decay data evaluation. Additional results on ^{97}Zr ($T_{1/2}$), ^{108}Ag and ^{110m}Ag (I_γ) data are foreseen. Isomeric branching ratios of ^{133}Cs(n,γ)^{134gs,m}Cs and ^{151}Eu(n,γ)^{152gs,m}Eu were also measured. Analyses are on-going.

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