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Abstract. The MAESTRO experimental program has been designed to improve nuclear data uncertainty on a large range of materials used for detection, absorption, moderation and structures in LWRs. It consists of pile-oscillation and neutron activation experiments, carried out in the MINERVE low power facility. For this program, the core configuration has been designed to be representative of HZP (Hot Zero Power) conditions of a typical PWR. Samples of high purity elements have been manufactured with severe technological constraints to reach a target accuracy of ±2% (1σ) on the measurement. This paper presents a preliminary analysis of activation experiments, based on TRIPOLI4 Monte-Carlo calculations and various nuclear data libraries.

1 Introduction

MAESTRO [1][2] is the follow-up in a series of experimental programs started more than thirty years ago in the MINERVE facility [3-4], which goal is to improve nuclear data uncertainties for Light Water Reactors (LWR) applications (GEN 2 and 3). Past programs mainly consist of pile-oscillation experiments on samples with small amount of separated isotopes, natural elements, fresh or irradiated fuels (UO_2, MOX).

Due to the lack of validation (or insufficient accuracy) for structural, moderating, detection and some absorbing materials, the MAESTRO experimental program was designed to improve the prediction of several neutronic parameters which are relevant for several LWR applications: core physics (reactivity of structural elements), instrumentation (EPR^{TM} innovative instruments), reactivity control, criticality/safety (contributions of various elements in concrete), burn-up credit. Some of the experiments are also performed in support of the JHR irradiation reactor, currently being built in Cadarache. This reactor will use metallic fuel plates of U3Si2 cladded with aluminum, a beryllium reflector and hafnium control rods.

This paper describes the experiments and preliminary results on the neutron activation measurements that have been performed on the following natural materials: Ag, In, Cs, Eu, V, Mo, Zn, Sn and Zr.

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2 Description of the experiment

2.1 MINERVE core configuration and related experimental techniques

MINERVE is a pool type reactor, located at CEA Cadarache. The core is submerged under 3 meters of light water at room temperature. The outside of the core is made of highly enriched UAl assemblies reflected by large graphite elements, being used as a driver zone. In a central square cavity of 70cm, various configurations can be loaded. For the MAESTRO program, a homogeneous core configuration consisting of ~800 UO_2 fuel pins (3% in ^{235}U) has been loaded. In the central position, the neutron spectrum is closed to the one of a UO_2-PWR in Hot Zero Power conditions (HZP).

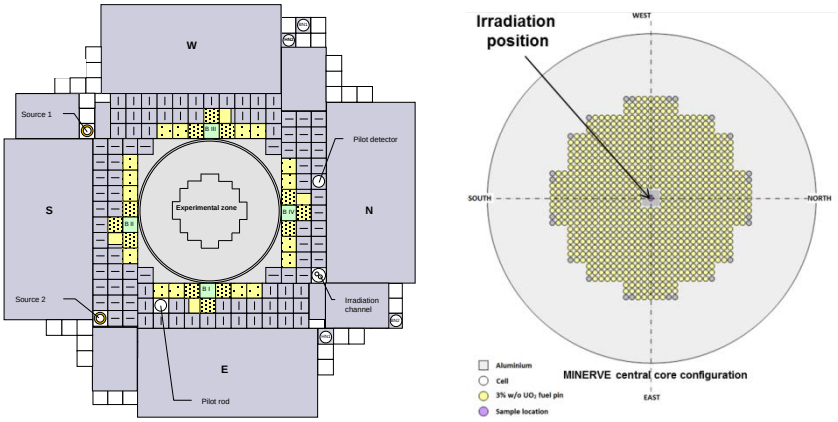

Figure 1. Overview of the MINERVE facility (left side) and central core configuration (right side)

Two kinds of experimental techniques have been used during the program. The first one is the pile-oscillation technique which consists in measuring the reactivity worth of a sample of pure material by successive translations between the center of the core and a position outside of the core. The technique is described in more details in [2] and [5]. The second one is the neutron activation techniques which consists in irradiating the sample in the center of the core and then to perform a γ-spectrometry analysis of the radionuclides which have been produced during the irradiation. This method was developed in previous programs to obtain complementary information on the capture rate of some absorbers and actinides, using the same sample in both pile-oscillation and activation techniques [6]. It is a relative method in which the reaction rate of a given isotope is normalized to the reaction rate of a reference isotope which is considered to be a neutron cross section standard.

2.2 Sample characteristics

All the samples used in MINERVE for the last fifty years consist of a mixture between a diluent material (DM) and a material of interest (MOI). For representativity considerations, UO_2 was often chosen as the DM. The MOI was mixed together with the DM, then crushed and sintered in pellets. Due to the possibility of material evaporation during the sintering process, additional pellets were prepared for destructive analysis by mass spectrometry. These analyses are quite expensive, time-consuming and can be complex for some insoluble elements.

For the MAESTRO program, the process was changed in order to avoid destructive analysis and to guarantee the material compositions just by accurate weighting and the provision of highly pure materials. Depending on physical and chemical characteristics of the samples, three different designs have been considered:

- Diluted solutions: this form uses a dilution of the MOI in 4-5% nitric acid. This form is well adapted for materials with high neutron cross section and for materials which are highly soluble. It is inserted in a double watertight cladding, one of welded Zircaloy-4 and one of aluminium alloy (Al-5754).
- Pure rods: this form uses cylinders of appropriated diameter for materials with low neutron cross section. The rod is inserted in a single watertight aluminium container.
- Powder mix: this form is a mixture between a DM (Al_2O_3 powder) and the MOI made of very fine powder (µm-sized powder). It is considered for materials which cannot be prepared from the two previous forms. The same cladding as for rod samples is used.

Table 1 summarises the characteristics of the samples used in the neutron activation experiments. Gold samples are used for calibration, as the capture of ^{197}Au is considered as a cross section standard.

Table 1. Characteristics of MAESTRO samples

Sample name	Material	Physical form	MOI mass (g)	Diameter (mm)	Height (mm)	Reaction of interest
M-Ag	Ag	4% HNO_3 solution	1.4	8.4	82	^{109}Ag(n,γ)^{110m}Ag
M-In	In	4% HNO_3 solution	0.22	8.4	82	^{113}In(n,γ)^{114m}In ^{115}In(n,γ)^{116m}In
M-Cs	Cs	4% HNO_3 solution	0.75	8.4	82	^{133}Cs(n,γ)$^{134gs+m}$Cs ^{133}Cs(n,γ)^{134m}Cs
M-Eu	Eu	5% HNO_3 solution	0.045	8.4	96	^{151}Eu(n,γ)^{152m}Eu ^{151}Eu(n,γ)^{152gs}Eu ^{153}Eu(n,γ)^{154}Eu
M-V	V	Powder mix	7.2	10	100	^{51}V(n,γ)^{52}V
M-Mo	Mo	Pure rod	29	6	100	^{98}Mo(n,γ)^{99}Mo ^{100}Mo(n,γ)^{101}Mo
M-Zn	Zn	Pure rod	52	10	100	^{64}Zn(n,γ)^{65}Zn ^{68}Zn(n,γ)^{69m}Zn
M-Sn	Sn	Pure rod	173	10	300	^{112}Sn(n,γ)^{113}Sn ^{117}Sn(n,γ)^{118m}Sn ^{122}Sn(n,γ)^{123m}Sn
M-Zr	Zy4 alloy	Pure rod	152	10	300	^{94}Zr(n,γ)^{95}Zr ^{96}Zr(n,γ)^{97}Zr
M-Au-1 M-Au-2 M-Au-3	Au	Pure rod	1.5 3.9 6.0	1 1.6 2	10	^{197}Au(n,γ)^{198}Au
M-AlAu	Al-0.1%Au alloy	Alloy rod	0.006	1	10	^{197}Au(n,γ)^{198}Au

2.3 Neutron activation experiments

The measurement procedure follows two steps. Firstly, the sample is introduced in a watertight stainless steel tube between aluminium spacers in order to have the sample at the radial and axial central position of the core. The reactor is set to an average power of 80W during 30 minutes to 6 hours, depending on the target activity to reach. Secondly, after waiting for a few minutes to a few hours (depending on the sample dose rate after the irradiation), the sample is placed in front of the HPGe detector to start a γ-spectrometry analysis (Fig. 2). This process is repeated at least twice for each sample in order to check the measurement reproducibility. For long-lived nuclides, a background subtraction is required to account for previous irradiations in the total activity.

In order to extrapolate the measured count rate to the saturated activity, the γ-counting rate is modified using several factors:
- the radioactive decay during the cooling and the measurement phases,
- the detector efficiency,
- the γ-emission probability of the γ-ray of interest.
A correction factor, called “efficiency transfer correction”, is also required to account for two effects:
- A spatial effect which is related to the volumic and heterogeneous distribution of the γ-source within the sample, while the detector efficiency was calibrated with a point source.
- An attenuation effect which accounts for the self-absorption within the sample, especially for the dense materials like gold.
This factor is evaluated by MCNP5 calculations (Fig. 2). More details can be found in [2] and [6].

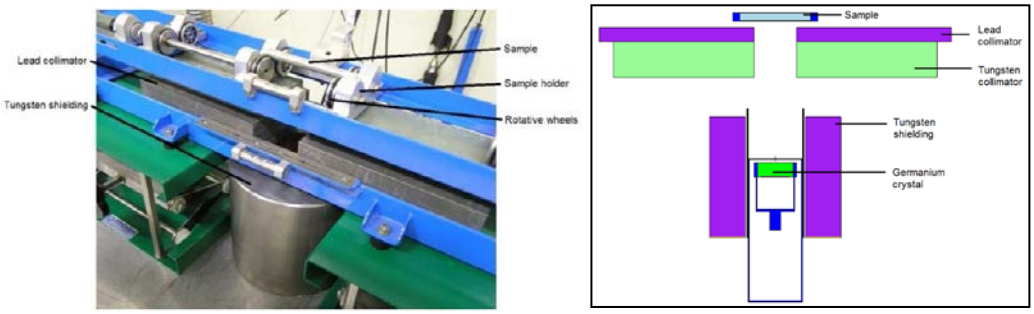

Figure 2. γ-spectrometry device (left side) and MCNP model for the efficiency transfer correction (right side)

The experimental results are presented as ratios of effective macroscopic capture rate relative to the gold macroscopic capture rate:

$$R_i = \frac{<N_i \sigma_{c,i}\, \phi>}{<N_{Au} \sigma_{c,Au}\, \phi>} \quad (1)$$

Four different gold samples were used and combined together. The combination of pure gold rods and aluminum-gold alloy wires is useful to validate the efficiency transfer correction between one with negligible (M-AlAu) and one with large (M-Au-3) self-absorption effect.

3 Interpretation of the neutron activation experiments

3.1 Calculation code and model

Neutron activation experiments were analysed by Monte-Carlo calculations based on TRIPOLI4 [7]. A detailed 3D model (Fig. 3) was settled including the graphite reflector, the driver zone (with the fuel assemblies explicitly described) and the experimental zone with the sample located in the core centre.

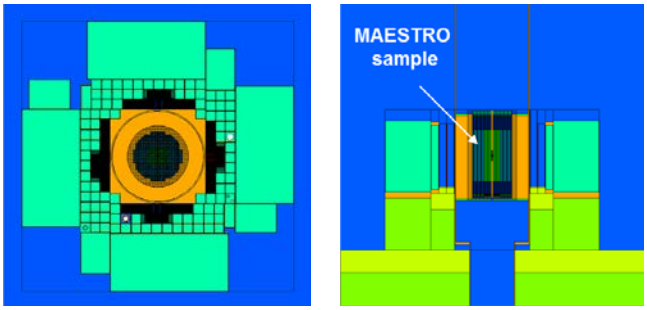

Figure 3. TRIPOLI4 3D detailed model of the MINERVE core

1 to 10 billion histories were simulated for each sample, leading to a convergence uncertainty of less than 0.5% (1σ). Three different neutron cross section libraries were considered for the calculation: JEFF-3.1.1 [8], JEFF-3.2 and ENDF/B-VII.0 [9]. As some reactions are partial captures, i.e. capture to a metastable stable, it was required to correct the computed total capture rate by the isomeric branching ratio. The value was taken from the EAF-2010 library [10] at thermal energy (25 meV).

3.3 Calculation vs Experiment

Table 2 presents the relative differences between calculated (C) and measured (E) capture rate ratios. The total uncertainty is a combination of the Monte-Carlo convergence (<0.5%), counting statistics (0.8 to 3%), technological uncertainty related to dimensions and compositions of the materials (<0.5%) and isotopic abundance in the natural element (0-3%).

Table 2. Calculation over measurement comparisons of macroscopic capture cross section ratios

Type/Interest of the material	Reaction	Total uncertainty (1σ)	TRIPOLI4.9 [C/E-1]		
			JEFF-3.1.1	JEFF-3.2	ENDF/B-VII.0
Control rod (AgInCd) and absorbing FP)	$^{109}Ag(n,\gamma)^{110m}Ag$	1.1%	0.0%	0.6%	-0.5%
Absorber (AgInCd)	$^{113}In(n,\gamma)^{114m}In$	2.0%	-12.6%	-12.1%	-11.4%
	$^{115}In(n,\gamma)^{116m}In$	1.6%	-3.3%	-2.7%	-3.7%
Absorbing Fission Product	$^{133}Cs(n,\gamma)^{134gs+m}Cs$	0.8%	-2.3%	-1.7%	-3.1%
	$^{133}Cs(n,\gamma)^{134m}Cs$	3.5%	-0.1%	0.5%	-0.9%
Absorber (possible burnable poison, Burn-up Credit)	$^{151}Eu(n,\gamma)^{152m1}Eu$	1.3%	-12.5%	-12.0%	-9.1%
	$^{151}Eu(n,\gamma)^{152gs}Eu$	2.1%	-12.1%	-11.6%	-8.7%
	$^{153}Eu(n,\gamma)^{154}Eu$	1.3%	-8.5%	-8.0%	-9.5%
Instrumentation	$^{51}V(n,\gamma)^{52}V$	1.3%	-1.4%	-0.8%	-2.2%
Structure (clads, grids)	$^{98}Mo(n,\gamma)^{99}Mo$	2.0%	-1.2%	-0.3%	-1.9%
	$^{100}Mo(n,\gamma)^{101}Mo$	3.5%	-0.5%	-3.2%	-1.3%
	$^{64}Zn(n,\gamma)^{65}Zn$	2.1%		2.3%	
	$^{68}Zn(n,\gamma)^{69m}Zn$	3.6%		7.0%	
Structure (Zy clads)	$^{112}Sn(n,\gamma)^{113}Sn$	2.0%	27.7%	28.8%	22.6%
	$^{117}Sn(n,\gamma)^{118m}Sn$	1.8%	-82.5%	-81.9%	-82.5%
	$^{122}Sn(n,\gamma)^{123m}Sn$	1.8%	18.7%	-8.3%	-11.7%
	$^{94}Zr(n,\gamma)^{95}Zr$	2.0%	17.3%	6.9%	17.0%
	$^{96}Zr(n,\gamma)^{97}Zr$	3.8%	-4.9%	-4.3%	-5.1%

Here are the conclusions that can be derived from this table:
- *Silver*: the C/E agreement is within 1σ uncertainty for the three nuclear data libraries. This trend is consistent with previously published results obtained from the BUC program in MINERVE, but with a significantly reduced uncertainty (from 4% to 1%) [11].

- *Indium*: the C/E agreement is within 2σ uncertainty for ^{115}In capture for the three libraries. All of them are also consistent with a 11-13% underestimation of ^{113}In capture. Additional results would be required to separate the contribution of the isomeric ratio to the capture cross section.

- *Cesium*: the C/E agreement is higher than 2σ uncertainty for ^{133}Cs total capture for the three libraries, with a systematic underestimation of 2-3%. The partial capture to ^{133m}Cs is predicted within 1σ, so it can be concluded than the isomeric ratio from EAF-2010 is correctly evaluated within 3.5%.

- *Europium*: the C/E agreement of the two ^{151}Eu partial capture is consistent for the three libraries, indicating an underestimation of 12% for JEFF-3.1.1 and JEFF-3.2 and 9% with ENDF/B-VII.0. Just like for caesium, one can conclude that the isomeric ratio to the first metastable state is correctly evaluated in EAF-2010. Moreover, it seems that a revision of the capture cross sections would be required to solve the underestimation of this capture rate ratio. Additional information will be provided by the analysis of the pile-oscillation experiments which will give information on the total capture rate of ^{nat}Eu (mostly depend on the one of ^{151}Eu). For ^{153}Eu, there is also an 8-9% capture cross section underestimation in the three libraries. Previous pile-oscillation experiments [10] and irradiation experiments [12] led to the same conclusions.

- *Vanadium*: the C/E agreement for ^{51}V capture is within 1σ for the JEFF libraries and within 2σ for the ENDF/B-VII.0. This trend confirms preliminary results previously published from the MAESTRO program, based on a pure rod sample [1].

- *Molybdenum*: the C/E agreement is within 2σ for both ^{98}Mo and ^{100}Mo capture cross section and for the three nuclear data libraries. These results complete the feedback obtained in the past on the ^{95}Mo and on the overall ^{nat}Mo captures [11].

- *Zinc*: no isotopic evaluations are available in JEFF-3.1.1 and ENDF/B-VII.0 so only the JEFF-3.2 can be computed. The C/E agreement is within 2σ for both ^{64}Zn and ^{68}Zn capture cross section.

- *Tin*: the C/E agreement for ^{112}Sn and ^{117}Sn capture is significantly outside of the 2σ uncertainty range. Further investigations are required to understand such large differences, reaching up to 80% for ^{117}Sn. For ^{122}Sn capture, the strong overestimation of 19% observed in JEFF-3.1.1 was reduced to -9% in JEFF-3.2 but it is still outside of the 2σ uncertainty, with the same trend for ENDF/B-VII.0. Additional work would be required to reach the 2σ uncertainty range.

- *Zirconium*: the C/E agreement for ^{96}Zr capture is within 2σ uncertainty for the three nuclear data libraries. For ^{94}Zr capture, a strong overestimation of 17% is observed with both JEFF-3.1.1 and ENDF/B-VII.0 but it is significantly reduced to 7% in JEFF-3.2. Nevertheless, additional work would be required to reach the 2σ uncertainty range.

4 Conclusions

The MAESTRO experiment benefits from the feedbacks of previous programs in the MINERVE reactor. A careful design of the samples with severe constraints on the material mass, purity and homogeneity, just like some improvements on the irradiation conditions, allows reaching a target accuracy of less than 2% on the experimental results (previously 3-4%).

Preliminary results on neutron activation experiments performed on natural elements were obtained with TRIPOLI4 Monte-Carlo calculations. They confirm JEFF-3.1.1 evaluations of ^{109}Ag, ^{115}In, ^{51}V, ^{98,100}Mo, ^{64,68}Zn and ^{96}Zr capture cross sections within 2σ uncertainty, while some other indicate that a possible re-evaluation should be required for ^{113}In, ^{133}Cs, ^{151,153}Eu, ^{112,117,122}Sn and ^{94}Zr. Moreover, it was possible to validate the isomeric ratio to the metastable state in the capture of ^{133}Cs and ^{151}Eu, thanks to the measurement of both ground and metastable states. All these results will have to be confirmed by the analysis of the pile-oscillation measurements that were performed on the same sample, which is currently ongoing.

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