EPJ Web of Conferences **284**, 01035 (2023) https://doi.org/10.1051/epjconf/202328401035
ND2022

Time-of-flight measurements of MINERVE samples containing fission products and neutron absorbing isotopes

Gilles Noguere[1,*], *Benoit* Geslot[1], *Adrien* Gruel[1], *Stefan* Kopecky[2]*Pierre* Leconte[1], *Carlos* Paradela[2], *Mathilde* Pottier[1], and *Peter* Schillebeeckx[2]

[1]CEA, DES, IRESNE Cadarache, F-13108 Saint Paul Les Durances, France.
[2]European Commission, Joint Research Centre, Geel, Belgium.

Abstract. The zero-power reactor MINERVE (CEA Cadarache) was designed to perform reactivity worth measurements by the oscillation technique. The various experimental programs, undertaken for the last thirty years, involved cylindrical samples with a diameter of about 1 cm and a height ranging from a few cm to 10 cm. Most of the samples are composed of UO_2 pellets mixed with a high neutron absorbing nuclide, i.e. fission product, actinide, in a double-sealed Zry-4 container. An experimental program started in 2015 in collaboration with the Joint Research Centre of Geel to study the MINERVE samples at the time-of-flight facility GELINA by the neutron transmission technique. The two main objectives consist of checking both the composition of the MINERVE samples provided by the manufacturer and the quality of the resonance parameters recommended in the evaluated neutron data library JEFF-3.3. The pioneer experiments on MINERVE samples containing ^{107}Ag and ^{109}Ag revealed a substantial Tungsten contamination that was not reported by the manufacturer. Such a Tungsten contamination is related to the manufacturing process of the sample pellets. The observed Tungsten contaminations lead to non-negligible increases of the C/E ratios up to a few percent. second experimental campaign on MINERVE samples containing ^{99}Tc provided useful insight on the quality of the ^{99}Tc resonance parameters measured at the GELINA facility at the end of the 90s. The ongoing program continuing through 2022 will deliver data for samarium (natural, 147, 149, 152), neodymium (natural,143,145), gadolinium (natural, 155), europium (151, 153), rhodium (103), cesium (133), hafnium (180), dysprosium (160, 161, 162, 163, 164) and erbium (168, 170). The present work focuses on the data analysis technique developed for long cylindrical samples with a diameter smaller than the neutron beam, and on the grain size distribution model implemented in the resonance shape analysis REFIT.

1 Introduction

Reactivity worth measurements by the oscillation technique in the MINERVE reactor of CEA Cadarache were performed with small cylindrical samples of 10 cm long. Since the shutdown of the facility, hundreds of samples containing actinides and non-fissile nuclides are now available for other type of experiments. The samples which contain an element enriched to a specific isotope mixed in a UO_2 matrix are well adapted to transmission measurements at the GELINA facility. The first phase of the program undertaken in collaboration with JRC-Geel and CEA of Cadarache deals with MINERVE samples containing non-fissile nuclides.

This paper briefly reminds the origin of the MINERVE samples (section 2), the formalism developed to analyze the transmission data measured at the GELINA facility (section 3) and the results obtained since the beginning of the experimental program (section 4).

2 MINERVE programs

The non fissile nuclides of interest for this study are part of the GADOLINIUM, Burn-Up Credit and OCEAN integral programs carried out at Cadarache in 1973, 1992 and 2005, respectively.

Burn-Up Credit (BUC) calculation routes account for the major actinides and fission products in criticality analyses for reprocessing applications and transport, storage and disposal of fuel assemblies. An experimental program between UKAEA (Winfrith) and CEA (Cadarache) was designed to validate the capture cross sections of 13 fission products in support to BUC studies, that encompassed the six main fission products that make up 50% of the anti-reactivity of all fission products: ^{103}Rh, ^{133}Cs, ^{143}Nd, ^{149}Sm, ^{152}Sm and ^{155}Gd. The goal of this program was to measure by the oscillation technique the reactivity worth of small samples composed of a stack of UO_2 pellets containing fission products in a double sealed zircaloy container. The experiments were performed in the DIMPLE (UKAEA) and MINERVE (CEA) reactors. Results are reported in Refs. [1, 2].

The integral trends delivered by the BUC program were complemented by the OCEAN program carried out

---

[*] e-mail: gilles.noguere@cea.fr

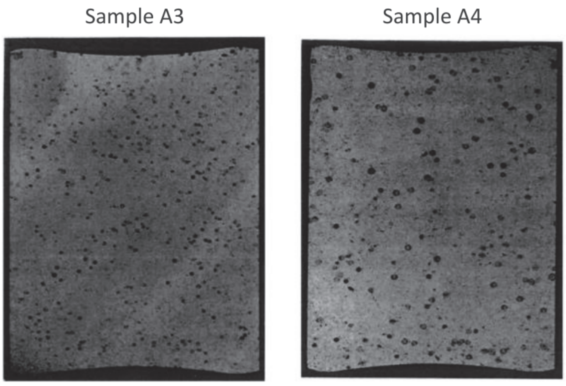

Figure 1. Micrograpgy of two UO_2 pellets containing microspheres of Gd_2O_3 of 120 and 195 μm.

in the MINERVE reactor only. This program was also designed to measure separately 16 nuclides with the aim of validating the capture cross sections of fission products not yet studied with MINERVE, but also of absorbant materials, such as hafnium, dysprosium and erbium. Analysis of the OCEAN results is still in progress. Obtaining final trends took time mainly because of contradictory sample compositions.

Due to the importance of the gadolinium isotopes as burnable poison, we also find an interest in samples that were measured in the framework of the GADOLINIUM program [3]. This wide program was designed to study enriched UO_2 pellets (5.1% ^{235}U) containing different amounts of gadolinium oxide (Gd_2O_3). Our study only focuses on experiments carried out in 1983 with samples containing microspheres of Gd_2O_3 of various diameters, ranging from 80 to 380 μm (Fig. 1).

Since 2010, a substantial effort was performed to accurately characterize samples used in the MINERVE programs by conventional chemical methods. In parallel, we decided to investigate the Neutron Resonance Transmission Analysis (NRTA) capabilities of the JRC-Geel as a non-destructive analysis technique for validating the composition of the MINERVE samples and testing resonance parameters recommended in the evaluated nuclear data libraries.

3 Governing equations

The transmission experiments were performed at the GELINA facility with the neutron detector positioned at 10 m from the neutron source. Figure 2 shows the geometry in which the MINERVE pellet samples were measured. For the analysis of such samples, special procedures were developed and validated at GELINA [4, 5]. The procedure proposed in this work is based on the one of Ref. [4]. It includes a dedicated formalism to account samples consisting of microspheres and a void fraction α. The experimental transmission T_{exp} as a function of time t can be deduced from the sample *in* and sample *out* measurements as follow:

$$T_{\text{exp}}(t) = \frac{[C_{in}(t) - B_{in}(t)] - \alpha[C_{out}(t) - B_{out}(t)]}{(1-\alpha)[C_{out}(t) - B_{out}(t)]}, \quad (1)$$

in which C and B represents the count rate and the background contribution, respectively. The corresponding theoretical transmission T as a function of the incident neutron energy E is given by:

$$T(E) = \frac{\int_{-r}^{r} \sqrt{1-\left(\frac{x}{R}\right)^2} \exp\left(-2r\sqrt{1-\left(\frac{x}{R}\right)^2} \sum_i \rho_i \sigma_{t,i}(E)\right) \mathrm{d}x}{\int_{-r}^{r} \sqrt{1-\left(\frac{x}{R}\right)^2} \mathrm{d}x}, \quad (2)$$

in which r is the sample radius, R is the beam radius, ρ_i defines the nuclide volume density and $\sigma_{t,i}$ stands for the total cross section of nuclide i. The geometrical relationship between the void fraction and the radii is:

$$\alpha = 1 - \frac{2R^2 \arcsin(r/R) + 2r\sqrt{R^2 - r^2}}{\pi R^2} \quad (3)$$

The presence of microspheres in the MINERVE samples can be approximated via the particle self-shielding correction f proposed by Doub [6]:

$$T_{\text{th}}(E) = T^f(E), \quad (4)$$

with

$$f = \frac{1}{\frac{2}{3}y\left(\frac{V}{g}\right)} \ln\left(\frac{1}{1-\left(\frac{V}{g}\right)(1-\bar{t})}\right), \quad (5)$$

in which V is a volume fraction that depends on the number of microspheres weighted by a compacting factor g. The collision probability $\bar{t}$ is derived from expressions given by Case [7]:

$$\bar{t} = \frac{2}{y^2}\left(1 - (1+y)e^{-y}\right), \quad (6)$$

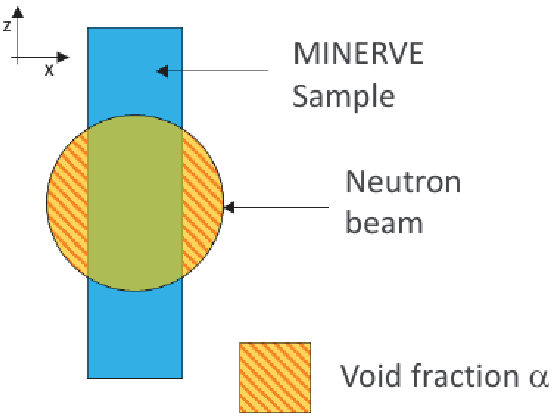

Figure 2. Drawing of a MINERVE sample placed in a neutron beam of diameter larger than the diameter of the sample.

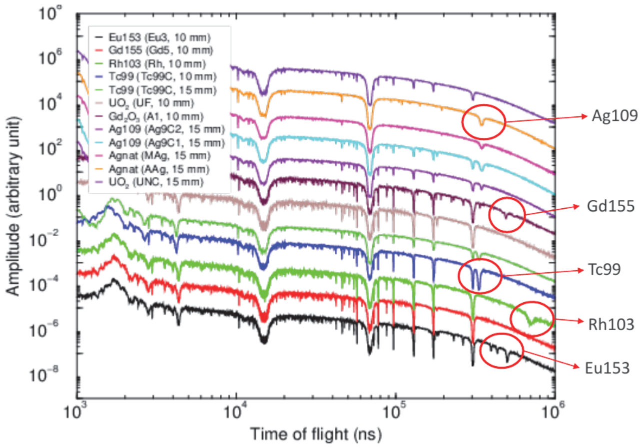

Figure 3. Examples of raw data obtained with MINERVE samples containing ^{107}Ag, ^{155}Gd, ^{99}Tc, ^{103}Rh and ^{153}Eu isotopes.

in which the intermediate parameter y depends on the volume density ρ_k and radius r_k of the microspheres k:

$$y = 2r_k\rho_k\sigma_{t,k}(E). \tag{7}$$

The equation (1) was used in the data reduction procedure through the AGS code [8], while Eqs. (2) to (7) were introduced in the resonance shape analysis code REFIT [9].

4 Results and discussions

The reduction of the raw data was performed with the AGS code. Figure 3 shows the various time-of-flight spectra obtained after dead time correction. The low energy resonances of silver, gadolinium, technetium, rhodium and europium can be well identified together with the contribution of the ^{238}U and black filter (Co and Na) resonances. Simple AGS command lines were combined to calculate the experimental transmission by taking into account the void fraction α. The latter void fraction was deduced from the bottom of the three first ^{238}U resonances, which are "black" in the case of the MINERVE samples.

The experimental transmission delivered by AGS and the theoretical transmission calculated with REFIT are compared in Fig. 4 for the MINERVE samples containing ^{155}Gd, ^{103}Rh and ^{153}Eu. The sample UF is a dummy (or reference) sample composed of a stack of UO_2 pellets, without fission product or neutron absorbing nuclide. In each plot, the resonance of ^{186}W highlights the non-negligible tungsten contamination, especially in the UF sample, coming from the fabrication process of the sample pellets. The tungsten contamination was first observed in MINERVE samples containing silver [4, 10] and ^{99}Tc [11]. In these cases the amount of tungsten ranges from 400 to 2800 ppm. The analysis of the ^{99}Tc sample also reveals the presence of 4066 ppm of Molybdenum. These surprising results indicate that GELINA is a suitable facility to accurately determinate the volume density of impurities in the MINERVE samples.

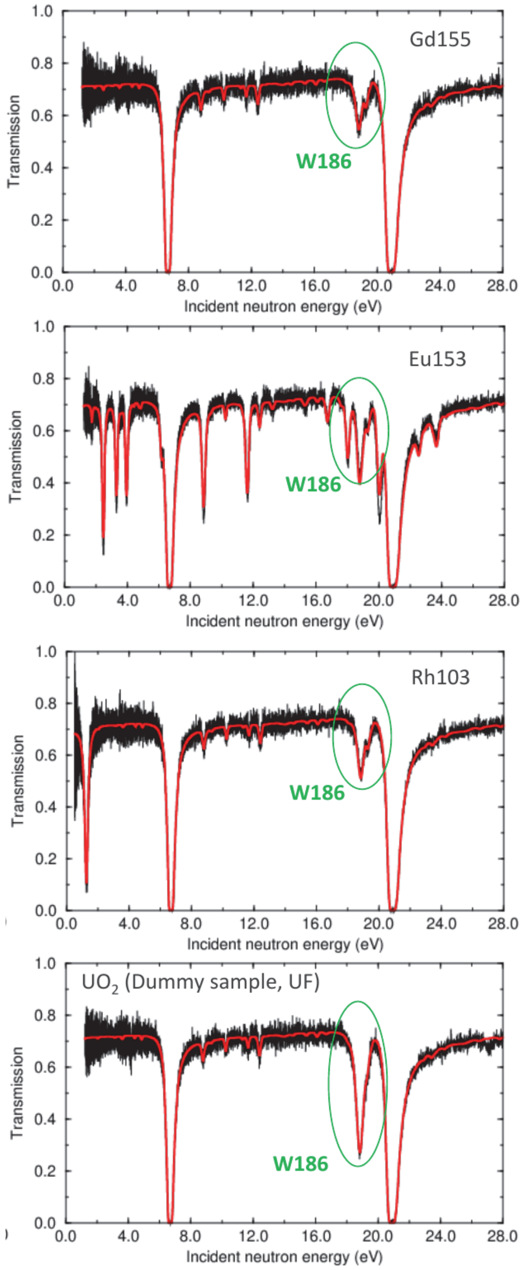

Figure 4. Examples of tungsten contamination observed in MINERVE samples. The solid curves represent the theoretical transmission calculated with the REFIT code.

The REFIT analysis of the transmission spectra measured with the MINERVE samples containing silver and technetium demonstrated that the results can be used to validate the resonance parameters recommended in the evaluated neutron data libraries. Our study provides capture resonance integrals equal to 99(3), 1514(22) and 314(12) b for ^{107}Ag, ^{109}Ag and ^{99}Tc, respectively. These results confirm the capture resonance integral of JEFF-3.1.1 for ^{109}Ag (1474.1 b) and ^{99}Tc (313.0 b), but suggests to decrease by −8.6% the capture resonance integral of ^{107}Ag (107.6 b). The corresponding resonance parameters are available in the test library JEFF-4T1.

For gadolinium, specific transmission measurements were carried out by using MINERVE samples composed

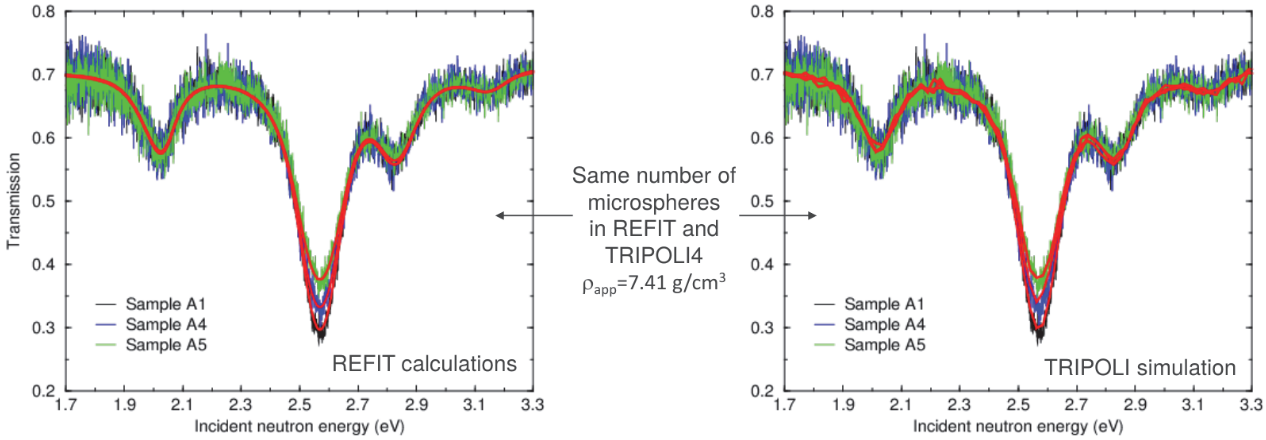

Figure 5. Examples of transmission obtained around the ^{155}Gd resonance at 2.6 eV by using MINERVE samples coming from the GADOLINIUM program. Each sample contains the same amount of Gd_2O_3. In sample A1, gadolinium oxide powder is homogenously mixed with the UO_2 matrix. In samples A4 and A5, the diameter of the microspheres of Gd_2O_3 is close to 195 and 380 μm, respectively. The same number of microspheres were used in the REFIT and Tripoli-4® calculations, which is equal to 6733 for sample A4 and 922 for sample A5.

of four enriched UO_2 pellets (5.1% ^{235}U) containing microspheres of Gd_2O_3. The aim of these transmission data is to validate the resonance parameters of gadolinium as well as to test the validity of the particle self-shielding correction proposed by Doub [6]. The equation (5) depends on the density of the microspheres. In this work, we considered the theoretical Gd_2O_3 density of 7.41 g/cm^3 to determine the number of microspheres seen by the neutron beam. Results obtained with REFIT around the ^{155}Gd resonance at 2.6 eV are shown in Fig. 5. In sample A1, gadolinium oxide powder is homogenously mixed with the UO_2 matrix. Therefore, the particle self-shielding correction was set to unity. In samples A4 and A5, the diameter of the microspheres of Gd_2O_3 is close to 195 and 380 μm, respectively. The particle self-shielding correction was calculated by introducing in the REFIT calculations 6733 microspheres for sample A4 and 922 microspheres for sample A5. The validity of these results was verified thanks to Monte-Carlo simulations performed with the Tripoli-4® code [12], in which 6733 and 922 microspheres were randomly generated in the volume intercepted by the neutron beam. The good agreement between the transmission data and the Tripoli-4® results confirms the validity of the Doub's model. Consequently, such a particle self-shielding correction could be conveniently introduced in the processing of the gadolinium evaluation to avoid excessive time calculations due to the individual description of the microspheres in Tripoli-4®.

5 Conclusions

The transmission experiments carried out at the GELINA facility with the MINERVE samples are useful for improving nuclear data libraries and the interpretation of past reactivity worth measurements performed at CEA of Cadarache. For example, in the case of the Burn-Up Credit program, results of the GELINA facility confirm the volume density of the main nuclides contain in the MINERVE samples and highlight the presence of impurities in the UO_2 matrix, such as tungsten and molybdenum. In the case of the GADOLINIUM program, the obtained results demonstrate that the particle self-shielding correction proposed by Doub can be used at the nuclear data processing level in order to avoid time-consuming Monte-Carlo calculations.

The experimental program on the MINERVE samples at the JRC-Geel is still in progress. It will delivers results for at least 30 non-fissile nuclides. The second phase of this program would concerned MINERVE samples containing actinides.

References

[1] A. Gruel et al., Nucl. Sci. Eng. 169, 229 (2011).
[2] C.J. Dean et al., Validation of important fission product evaluations through CERES integral benchmarks, in Proc. Int. Conf. Nuclear Data for Science and Technology, Nice, France, 2007.
[3] P. Chaucheprat, Qualification du calcul des poisons consomables au gadolinium dans les REL, PhD Thesis, Paris-Sud University, 1988.
[4] L. Salamon et al. J. Radio. Nucl. Chem. 321, 519 (2019).
[5] Ma et al. J. Anal. At. Spectrom. 35, 478 (2020).
[6] W.B. Doub, Nucl. Sci. Eng. 10, 299 (1961).
[7] K.M. Case et al., Introduction to the theory of neutron diffusion, Los Alamos Scientific Laboratory, 1953.
[8] B. Becker et al., J. Instrum. 7, 11002 (2012).
[9] M.C. Moxon and J.B. Brisland, REFIT: a least squares fitting program for resonance analysis, Report AEA Technology, AEA-InTec-0630, 1991.
[10] L. Salamon et al., Nucl. Instrum. Method B 446, 19 (2019).
[11] G. Noguere et al., Phys. Rev. C 102, 015807 (2020).
[12] E. Brun et al., Ann. Nucl. Energy 65, 151 (2015).