

OCEAN : an Ambitious Experimental Program for the Qualification of Integral Capture Cross Sections of Neutron Absorbers

Jean-Pascal Hudelot^{*}, Muriel Antony, David Bernard, Pierre Leconte, Serge Testanière,
Philippe Fougeras

*Commissariat à l'énergie atomique, Nuclear Energy Division,
13115 Saint-Paul-Lez-Durance Cedex, FRANCE*

Abstract

OCEAN (Oscillation in Core of SamplEs of Neutron Absorbers) is an ambitious experimental program supported by EDF and CEA. It is carried out in the MINERVE reactor of CEA Cadarache which is a low-power uranium fueled pool reactor. It started in 2005 and will end in 2008.

It aims at improving the knowledge on nuclear basic data for the neutron absorbers. It deals at the same time with the improvement of the calculation tools and with the feasibility studies on the new options of the fuel cycle. It particularly supports, for the LWR reactors, the studies on the increase of the fuel cycle length in nuclear power plants, and on the plutonium management. It also aims at qualifying the calculation tools as regard with new neutron absorbers. The main framework is the European JEFF3 project.

More in details, it deals with providing precise experimental data (capture cross sections) about the following absorbers : Gd-155, Gd-157, Gd-Nat, Hf-177, Hf-178, Hf-179, Hf-180, Er-166, Er-167, Er-168, Er-170, Dy-160, Dy-161, Dy-162, Dy-163, Dy-164, Eu-151, Eu-nat, and Eu-153.

The study of the neutron absorbers is performed on a large range of neutron spectra (over-moderated thermal spectrum, PWR UO_x standard spectrum, PWR 100% MOX spectrum and epithermal HCLWR type spectrum) corresponding respectively to the R2-UO₂, R1-UO₂, R1-MOX and MORGANE-R experimental lattices inside the MINERVE reactor.

This paper first presents the objectives of the OCEAN experimental program. Then the description of the MINERVE facility is given, focusing on the different core configurations that will be studied, and the oscillation technique is reminded. Besides, the oscillation samples specially fabricated for the OCEAN program are described. Finally, the first preliminary experimental results in the R1-UO₂ lattice are given and commented.

KEYWORDS: *OCEAN samples, absorbers, oscillation technique, reactivity worth, spectral indices, integral cross sections, MINERVE*

1. Introduction

OCEAN (Oscillation in Core of SamplEs of Neutron Absorbers) is an ambitious experimental program supported by EDF and CEA. It is carried out in the MINERVE reactor

^{*} Corresponding author, Tel. +334 42 25 43 58, Fax. +334 42 25 78 76, E-mail: jean-pascal.hudelot@cea.fr

of CEA Cadarache which is a low-power uranium fueled pool reactor [1]. It started in 2005 and will end in 2008.

It aims at improving the knowledge on nuclear basic data for the neutron absorbers. It deals at the same time with the improvement of the calculation tools and with the feasibility studies on the new options of the fuel cycle. It particularly supports, for the LWR reactors, the studies on the increase of the fuel cycle length in nuclear power plants, and on the plutonium management. It also aims at qualifying the calculation tools as regard with new neutron absorbers. The main framework is the European JEFF3 project.

First the objectives of the OCEAN experimental program are detailed. Then the description of the MINERVE facility is given, focusing on the different core configurations that will be studied, and the oscillation technique is reminded. Besides, the oscillation samples specially fabricated for the OCEAN program are described. Finally, the first preliminary experimental results on conversion ratio and reactivity worth measurements in the R1-UO₂ lattice are given and commented.

2. Objectives of the OCEAN experimental program

The need of better nuclear data on neutron absorbers and poisons becomes more and more important. It especially deals with the increase of burn up in PWRs, the studies on plutonium management and also the future concepts of reactors. The precise knowledge of these nuclear data is a major stake for the validation of calculation tools and thus for the prediction of the behaviour of those isotopes in different core configurations.

Considering the objective to increase burn ups, it needs to increase the U-235 enrichment of the fuel. Consequently it becomes necessary to raise the control of the reactor in order to compensate the initial excess of reactivity of the core. As it is not possible to increase the boron concentration (due to chemical issues in the primary circuit and to keep a negative moderator temperature coefficient), the use of new burnable poisons has to be envisaged.

Dealing with the plutonium management, advanced concepts based on full MOX loading involve a decrease of the efficiency of the control systems and a degradation of the moderator temperature coefficient. New and more efficient absorbers are thus necessary to be developed to get rid of these issues.

In this framework, the OCEAN program aims at providing an exhaustive experimental database on the integral capture rates of separated neutron absorbers and burnable poisons (Gadolinium, Erbium, Dysprosium, Europium and Hafnium), in a wide range of neutron spectra (from over-moderated to epithermal spectra).

More in details (see Tab. 1), it deals with providing precise experimental data (capture cross sections) about the 19 following absorbers : Gd-155, Gd-157, Gd-Nat, Hf-177, Hf-178, Hf-179, Hf-180, Er-166, Er-167, Er-168, Er-170, Dy-160, Dy-161, Dy-162, Dy-163, Dy-164, Eu-151, Eu-nat, and Eu-153.

3. The MINERVE facility

3.1 General description of the MINERVE facility

MINERVE is a pool type reactor operating at a maximum power of 100 W. The core is submerged under 3 meters of water and is used as a driver zone for the different experiments located in a central cavity with a size of about 70 cm x 70 cm. The coupled lattices are built so that they can reproduce the neutron spectrum of a water reactor (thermal or epithermal) or of a fast reactor [1] [2].

Table 1: Target improvements on nuclear data of neutron absorbers from the OCEAN Program

Isotope	current uncertainties (%)		target uncertainties (%)
Gd-155	$\sigma_{a\text{therm}}$ (a)	4%	2%
	I_R (b)	7%	3%
Gd-157	$\sigma_{a\text{therm}}$ (a)	3%	1.5%
	I_R (b)	6%	4%
Hf-177	$\sigma_{a\text{therm}}$	3%	1.5%
	I_R (b)	5%	3%
Hf-178	I_R (b)	7%	4%
Hf-179	I_R (b)	10%	4%
Hf-180	$\sigma_{a\text{therm}}$ (a)	3%	2%
	I_R (b)	5%	3%
Er-166	$\sigma_{a\text{therm}}$ (a)	30%	3%
	I_R (b)	30%	4%
Er-167	$\sigma_{a\text{therm}}$ (a)	4%	2%
	I_R (b)	13%	4%
Er-168	I_R (b)	13%	5%
Er-170	I_R (b)	10%	4%
Dy-160	I_R (b)	11%	4%
Dy-161	$\sigma_{a\text{therm}}$ (a)	5%	2%
	I_R (b)	9%	4%
Dy-162	$\sigma_{a\text{therm}}$ (a)	5%	3%
	I_R (b)	10%	3%
Dy-163	$\sigma_{a\text{therm}}$ (a)	6%	3%
	I_R (b)	7%	3%
Dy-164	$\sigma_{a\text{therm}}$ (a)	5%	2%
	I_R (b)	6%	4%
Eu-151	$\sigma_{a\text{therm}}$ (a)	4%	2%
	I_R (b)	10%	4%
Eu-153	$\sigma_{a\text{therm}}$ (a)	5%	3%
	I_R (b)	7%	3%

(a) I_R = resonance integral

(b) $\sigma_{a\text{therm}}$ = thermal absorption cross section

3.2 The different core configurations

The study of the neutron absorbers is performed on a large range of neutron spectra (over-moderated thermal spectrum, PWR UOx standard spectrum, PWR 100% MOX spectrum and epithermal HCLWR type spectrum) corresponding respectively to the R1-UO2 (see Fig. 1), R1-MOX (see Fig. 2), R2-UO2 (see Fig. 3) and MORGANE-R (see Fig. 4) experimental lattices inside the MINERVE reactor.

Figure 1 : R1-UO2 lattice
(PWR spectrum)

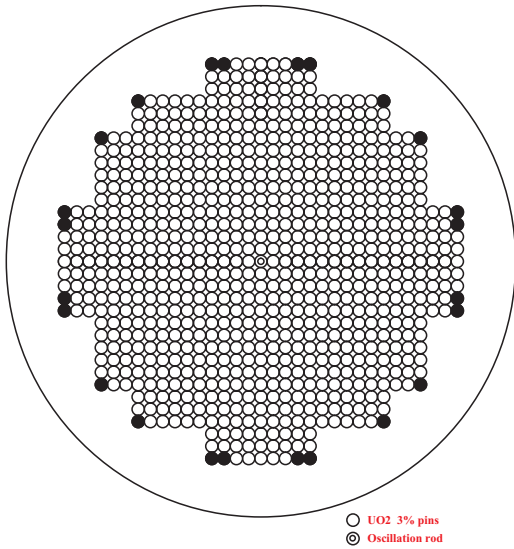


Figure 2 : R1-MOX lattice
(PWR-MOX spectrum)

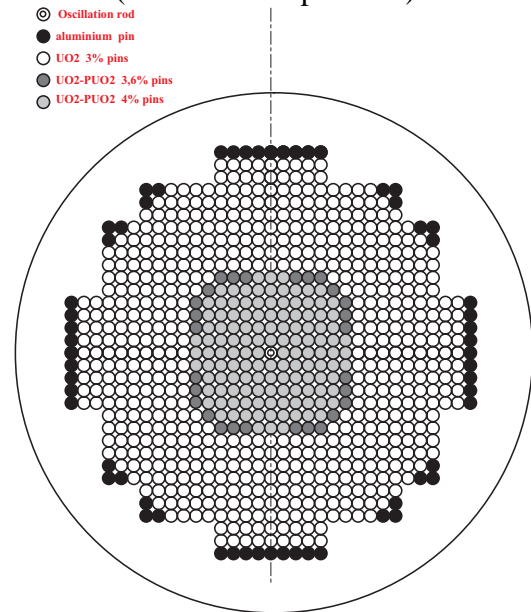


Figure 3: R2-UO2 lattice
(over-moderated spectrum)

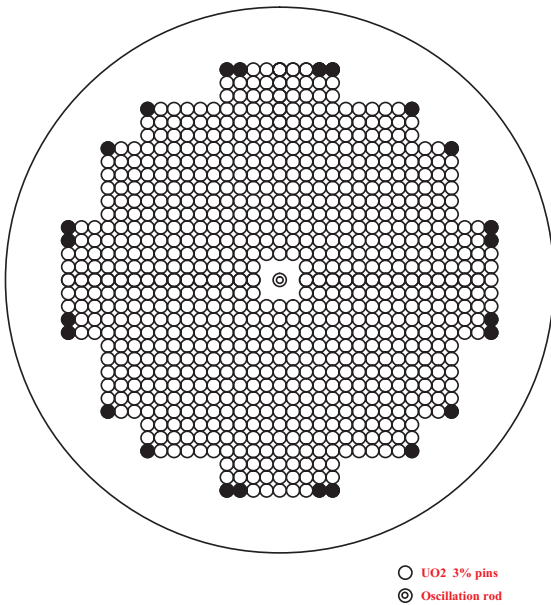
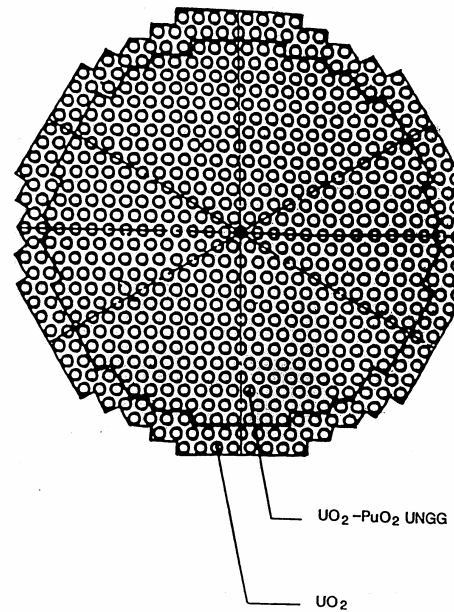


Figure 4: MORGANE-R lattice
(HCLWR spectrum)



4. The sample fabrication and chemical analysis

4.1 The sample fabrication

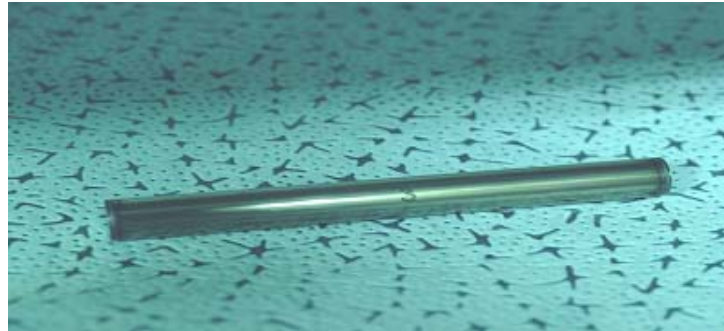
The samples consist of a doping absorber homogeneously mixed inside a sintered natural UO_2 matrix. They are fabricated in the Fuel Studies Department (DEC) of CEA Cadarache. The column of fuel pellets has a diameter of 8.1 mm and is 95 mm high. It is inserted in a

double welded and waterproof Zircaloy4 clad, with a overall diameter and height of 10.6 mm and 103.5 mm.

A special attention was given to the homogeneity of repartition of the doping isotopes inside the UO_2 matrix. Precise specifications were respected about the density of fuel (about 95%) and about the standard deviation on the diameter and the height of the columns of fuel pellets.

An example of OCEAN fuel sample is given in Fig. 5.

Figure 5: example of OCEAN oscillation sample



4.2 The chemical analysis

High accuracy chemical and isotopic analysis are performed to control as well as possible the material balance of the samples, and thus to reduce the contribution of the material balance to the uncertainty on the results.

In order to avoid systematic errors on the analysis due for example to the calibration of the analytic tools, the chemical analysis are performed in 2 different laboratories of CEA Marcoule and CEA Saclay.

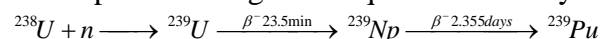
5. The measurement program

5.1 Characterization of the neutron spectrum

Before using the calculation tools for analyzing the oscillation measurement results, it is necessary to check that they are able to well reproduce the neutron spectrum at the position of the fuel samples. In this frame, conversion ratio and spectral indices measurements were performed in the R1-UO2 lattice.

5.1.1 Conversion ratio measurements by gamma-spectroscopy

The conversion ratio, noted $C8/F_{tot}$, is the ratio of neutron capture in ^{238}U compared to the total fission rate. It thus gives an information on the neutron spectrum. It is obtained via gamma spectroscopy measurements of fuel pins in the experimental region. The principle of the γ -ray spectroscopy measurements is to determine the reaction rate of a nuclide by measuring its specific activity. The determination [3] [4] is based on the measurement of the integral photopeak of a high-yield fission product relative to the total fission rate inside the fuel pin. The specific fission product gamma ray line is the 293.27 keV line from Ce-143. The 277.60 keV gamma ray line from ^{239}Np is used to measure the ^{238}U capture rate because it is related to the number of ^{238}U captures through subsequent beta decays:



5.1.2 Spectral indices by gamma-spectroscopy

A technique for measuring the spectral indices $C_X(n,\gamma) / F_{tot}$, defined as the ratio of the (n,γ) capture rate on “X” to the total fission rate, has been developed and improved, based on the modified conversion ratio measurement technique (see § 5.1.1). The measurement consists of γ -ray spectrometry applied directly on irradiated fuel samples, in order to determine the capture and fission rates with their specific activities. The feasibility of the experiment depends on some properties of the reaction products, as the radioactive periods, the γ -ray emission probabilities and the fission yields.

The adapted isotopes for this technique are: Dy-164, Er-170, Hf-180, Eu-151 and Eu-153. The different steps of the measurement are the net peak area measurements, the decay and dead time corrections, the experimental efficiency calibration of the Low-Energy Germanium detector, the calculation of efficiency transfers and of the average fission yields.

More details about the experimental technique can be found in reference [5].

5.2 Reactivity worth of the OCEAN samples using the oscillation technique

The technique consists in oscillating samples that contain the studied absorber in the center of the experimental lattice in order to measure the associated reactivity variation with an accuracy better than 1% (at 1σ). Each sample is placed in an oscillation rod and moved periodically and vertically between two positions located in and out of the experimental zone as shown in Fig. 6.

The studied sample is compared to a reference sample that differs just by the lack of absorber and that is placed in the bottom of the oscillation rod. Each sample is measured at least 5 times in order to significantly decrease systematic errors. A measurement corresponds to 10 oscillations of 120 seconds each.

The variations of flux induced by the oscillation are detected by a fission chamber placed in the driver zone, called the pilot chamber, that is servo-driven to a rotary automatic pilot rod. The pilot rod uses cadmium sectors, as shown in Fig. 7, to compensate for the reactivity variations. It is calibrated using U-235 and B-10 samples, whose reactivity worth is known with uncertainties better than 1% through deterministic calculations.

Figure 6: Movement of the oscillation sample inside the MINERE facility

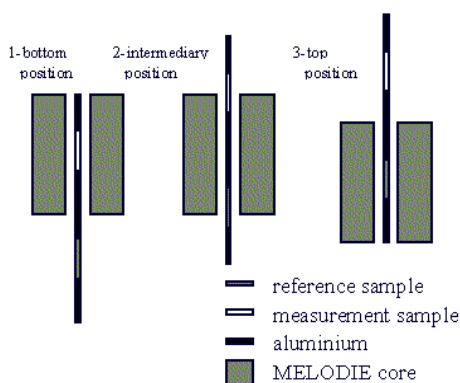
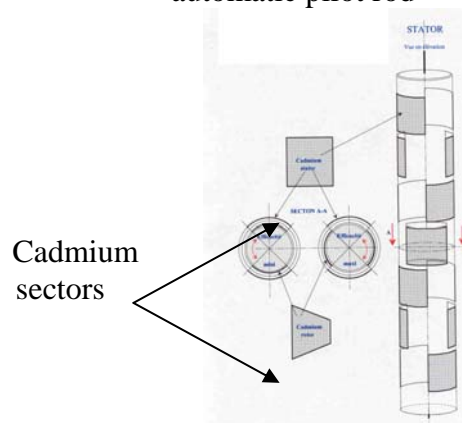


Figure 7: Scheme of the MINERVE automatic pilot rod



Taking into account the uncertainties on the measurement ($\sim 1\%$), the material balance of the samples ($\sim 2\%$), and the calibration of the pilot rod ($\sim 2\%$), the final experimental accuracy on the reactivity worth is about 3% (at 1σ). More details about the oscillation technique, the experimental data analysis and the associated uncertainties can be found in [2] and [6].

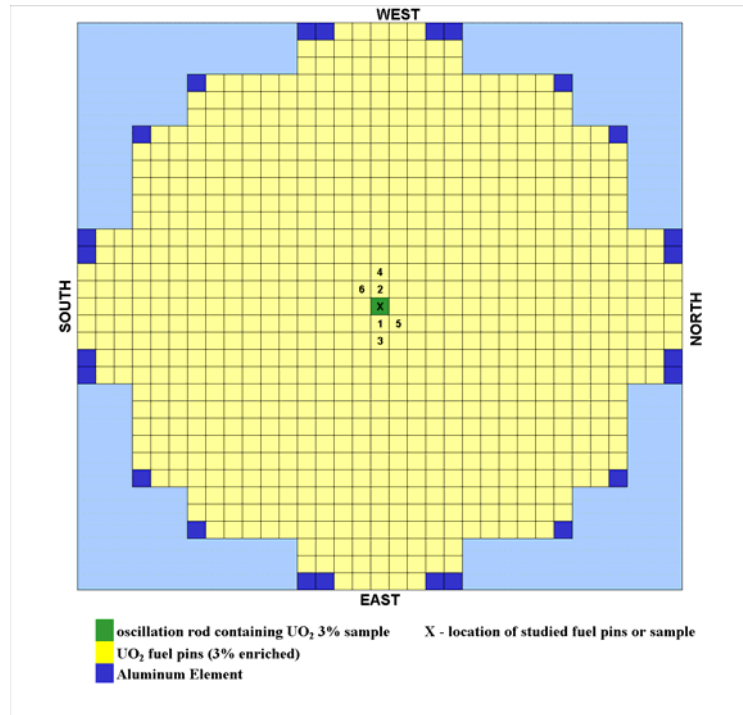
6. Preliminary experimental results – Comparison to calculational results

6.1 Characterization of the neutron spectrum

6.1.1 Conversion ratio measurements by gamma-spectroscopy

The conversion ratios were measured in the R1-UO₂ lattice, on 6 fuel pins around the central cell, and on a UO₂ sample enriched at 3% in U-235 placed in the center (see Fig. 8).

Figure 8 : location of the studied fuel pins and sample for the conversion ratio measurements in R1-UO₂



Tab. 2 gives the experimental results, and the comparison with 172 energy group deterministic APOLLO2 code calculations [6] [7], made with the JEFF3.1 data library. The experimental uncertainty, about 3.5 % in every case, is the combination of the uncertainty of reproducibility of the experiment (about 0.5%) and of the uncertainty (about 3.4%) due to the basic nuclear data (radioactive periods, gamma emission intensities, fission yields) used for treating raw measurement data.

An excellent agreement, within 1σ of uncertainty, between calculational and experimental results is shown, so that it represents a first validation of the calculation scheme used for analysing the oscillation measurements.

Table 2 : Comparison of conversion ratio experimental results with calculational results

Fuel pin or sample	Calculation / Experiment (C/E)	
	C/E	$\sigma(E)/E$ (a)
1	0.998	3.49%
2	1.009	3.49%
3	1.007	3.49%
4	1.000	3.48%
5	1.007	3.49%
6	1.001	3.48%
UO ₂ 3% sample	1.027	3.46%

(a) relative standard deviation on experiments

6.1.2 Spectral indices by gamma-spectroscopy

The spectral indices (see § 5.1.2) were measured on the Dy-164, Er-170, Hf-180, Eu-151 and Eu-153 samples, placed in the center of the R1-UO2 lattice, inside an oscillation rod.

The spectral indices measurement results are given in Tab. 3 and compared with reference continuous energy probabilistic MCNP code calculations and with 172 energy group deterministic APOLLO2 code calculations, made with the JEFF3.1 data libraries [5].

Table 3 : Comparison of spectral indices experimental results with calculational results

sample	Calculation / Experiment (C/E)			
	C/E with the APOLLO2 code + JEFF3.1 library	$\frac{\sigma(C/E)}{C/E}$ (a)	C/E with the MCNP4C2 code + JEFF3.1 library	$\frac{\sigma(C/E)}{C/E}$ (a)
Dy-164	0.866	10.9%	0.853	10.9%
Er-170	0.839	4.31%	0.812	4.40%
Hf-180	0.881	1.47%	0.865	2.48%
Eu-151	1.322	1.14%	1.361	1.45%
Eu-153	0.95	1.21%	0.985	1.49%

(a) combined uncertainty of the calculation and the experiment

Tab. 3 shows that the experimental and the calculational results are not in agreement within the uncertainties (2σ), save in the case of Eu-153. However, the results obtained with the APOLLO2 model are consistent at 1σ with the ones obtained with the reference MCNP model, so that the discrepancies between experiments and calculations can be assumed as coming from the nuclear data (gamma emission probabilities and/or (n, γ) cross sections) of the studied isotopes in JEFF3.1.

The calculation with the JEFF3.1 data library over-estimates by about 30% the spectral index related to Eu-151. For Eu-153, a rather good consistency is found between calculational results and experimental results.

The cases of Dy-164, Er-170 and Hf-180 are discussed in section 7.

The comparison of the spectral indices C/Es with the oscillation results should allow in the future to identify and quantify whether the discrepancies between calculated and measured spectral indices most likely come from the gamma emission probabilities or the (n, γ) cross sections of the studied absorbers.

6.2 Reactivity worth of the OCEAN samples using the oscillation technique

The OCEAN samples were oscillated in the central cell of the R1-UO2 lattice between October 2005 and May 2006. In most of the cases, the relative experimental uncertainty was about 1% (at 1σ), so that it was consistent with the predictions and the objectives of the experimental program in term of precision. Tab. 4 gives the preliminary (C/E) comparison of calculational results with experimental results.

The calculations were performed using the APOLLO2 deterministic code in a 2D model, with 172 energy groups and space-dependant self shielding, using 2D interface currents with imposed leakage. Both JEF2.2 and JEFF3.1 data libraries were considered.

As only few chemical analysis on the samples were at that time performed on the following samples – Hf-180, Er-170 and Dy-164, the preliminary C/E are only given for those samples.

Furthermore, due to temporary issues on the calibration of the signal, the uncertainty has been over-estimated in consequence, which explains the overall uncertainties on C/Es

between 5% and 15%. After treatment of this issue, the overall uncertainties on all the C/E should be likely around 5%.

Table 4 : Comparison of oscillation experimental results with calculational results, in the R1-UO₂ lattice of the MINERVE facility

Sample	$\frac{\sigma(E)}{E}$ (a)	Calculation / Experiment (C/E) with JEFF3.1	$\frac{\sigma(C/E)}{C/E}$ (b)
Hf-180	2.7%	0.751	10%
Er-170	2.6%	0.629	15%
Dy-164	0.6%	0.817	5%

(a) uncertainty on the reproducibility of the oscillation measurement (at 1σ)

(b) combined uncertainty on the data analysis, the material balance of samples and the experiment

7 Discussion and conclusion

Despite the current rather high uncertainties on the results, these last ones show the same tendencies as the ones obtained with the spectral indices measurements. Moreover, the oscillation measurement and the spectral indices measurements are consistent with 1σ of the total uncertainty.

As a conclusion, it is observed that the integral radiative capture cross sections of Hf-180 and Dy-164 in R1-UO₂ tend to be underestimated of about 20% in JEFF3.1, and the integral radiative capture cross section of Er-170 is over-moderated of about 25%.

In the near future, lower uncertainties (about 5%) will be carried out on the oscillation measurements, allowing to better quantify the tendencies on capture cross sections. Besides, results will be soon available for all the other studied neutron absorbers.

Moreover, the comparison of the spectral indices results with the oscillation results should allow in the future to identify and quantify whether the discrepancies between calculated and measured spectral indices most likely come from the gamma emission probabilities or the (n,γ) cross sections of the studied absorbers.

Finally, similar spectral indices and oscillation measurement are expected to be performed in other types of neutron spectra (over-moderated, PWR-MOX and RSM-epithermal spectra) by the end of 2008. The combination of the results obtained in the different configurations will permit to identify whether the discrepancies between calculation and experiments come from the thermal region or from the resonance region of the radiative cross sections of the studied absorbers.

Acknowledgements

The authors of this paper thank all the participants to the experimental program and their industrial partner Electricité de France (EDF) for their financial support.

References

- 1) P. Fougeras, JP. Hudelot, M. Antony, F. Mellier, C. Destouches, P. Blaise, S. Cathalau, N. Thiollay, "Eole, Minerve and Masurca facilities and their associated neutron experimental programs", ICONE 13, 13th International Conference on Nuclear Engineering, May 16-20, 2005, Beijing, CHINA
- 2) JP. Hudelot, R.T. Klann, P. Fougeras, F. Jorion, N. Drin, L. Donnet, "OSMOSE: An Experimental Program for the Qualification of Integral Cross Sections of Actinides", PHYSOR 2004 Conference, April 25-29, 2004, Chicago, IL USA

- 3) JP. Hudelot, S. Cathalau, JP. Chauvin, P. Fougeras, V. Laval, H. Philibert, "Measurement of the Modified Conversion Ratio of ^{238}U and of the ^{242}Pu Capture Rate by Gamma-Ray Spectroscopy on Irradiated Fuel Pins", Eleventh International Symposium on Capture Gamma-Ray Spectroscopy, Pruhonice, Czech Republic, September 2 - 6, 2002
- 4) R. Klann, JP. Hudelot, M. Antony, B. Micklich, G. Perret, N. Thiollay, G. Imel, JM. Girard, V. Laval, "MINERVE Reactor Characterization in Support of the OSMOSE Program: Spectral Indices", PHYSOR 2004 Conference, April 25-29, 2004, Chicago, IL USA
- 5) P. Leconte, JP. Hudelot, M. Antony, "Integral capture rate measurements of minor actinides by γ -ray spectrometry on specific fuel samples", PHYSOR 2006 Conference, 2006 September 10-14, Vancouver, BC, Canada
- 6) JP. Hudelot, R.T. Klann, D. Bernard, M. Antony, P. Fougeras, F. Jorion, N. Drin, L. Donnet, C. Leorier, "Preliminary Experimental Results of the OSMOSE Program for the Qualification of Integral Cross Sections of Actinides", PHYSOR 2006 Conference, 2006 September 10-14, Vancouver, BC, Canada
- 7) D. Bernard, A. Courcelle, O. Litaize, A. Santamarina, M. Antony, JP. Hudelot, "Validation of Actinide Neutron Induced Cross-sections: Preliminary Analysis of the OSMOSE experiment in MINERVE", PHYSOR 2006 Conference, 2006 September 10-14, Vancouver, BC, Canada