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Measurement of gamma-ray emission probabilities using a combination of activation and oscillation techniques: applications to ^{171}Er, ^{181}Hf, ^{233}Pa and ^{243}Pu

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Abstract. Emission probabilities of γ-rays are one of the most important decay parameters and reduction of their uncertainty is required in many fields of nuclear physics. For some radionuclides, a direct measurement of these parameters is not achievable with a sufficient accuracy due to the limitations of the usual experimental techniques. In this paper, an innovating method for measuring some γ-ray emission probabilities is proposed. It is based on the combination of oscillation and activation techniques. On one hand, the oscillation measurements provide an accurate estimation of the integral absorption cross sections of the studied isotope. On the other hand, the integral capture cross sections are also determined by γ-ray spectrometry using the relevant γ-rays of the capture product. The comparison of those independent results gives a direct and accurate assessment of the γ-ray emission probabilities. The combined method is fully described, as well for instruments and conditions as for the control of the uncertainties. It is applied to the cases of ^{171}Er, ^{181}Hf, ^{233}Pa and ^{243}Pu. The study enlightens an overestimation of (19.1 ± 20.5)%, (28.6 ± 12.7)% and (20.0 ± 12.3)% of the JEFF3.1 emission probabilities of the most intense γ-ray of respectively ^{171}Er, ^{181}Hf and ^{243}Pu. In the near future, this technique will be applied to ^{238}Np and ^{244}Am.

1 Introduction

One of the requirements for the quantitative estimation of radionuclides by means of γ-spectrometry is the knowledge of the relevant γ-rays. It also deals with the control of the decay-heat power and with the accuracy of reaction rate measurements – using γ-ray spectrometry. Besides, the GEN IV concepts of reactor, the increase of the fuel cycle length and the waste management in the current nuclear power plants, need to take into account the increasing amounts of minor actinides and the use of new neutron absorbers. Consequently, the knowledge of the emission probabilities for their relevant γ-rays has to be improved.

By the past, γ-ray emission probabilities were usually evaluated from the β-ray branching ratios and the relative γ-ray intensities because they could not be measured directly. These measurements were limited by the insufficient accuracy of the β-ray branching ratios, especially because of the inability to detect weak high-energy γ-ray emissions satisfactory, which impacts on the β^- decay data (the so-called "pandemonium effect" [1]).

Later, $4\pi\beta$-γ coincidence systems [2] were developed to measure the absolute disintegration rate without using the β-ray branching ratios while the absolute γ-ray intensities were obtained using a HPGe detector. Significant improvements in the knowledge of decay scheme were obtained from this technique. Nevertheless, the accuracy of the measurement hardly depends on the purity of the sample which implies a well known chemical separation of the isotopes, because no discrimination can be done on the measured coincidences.

To solve some of these limitations, an innovative technique was built up to deduce the absolute disintegration rate from the reaction rate on the parent isotope. As the accuracy of this experiment is directly linked to the knowledge of the capture cross section, pile oscillation measurements were made in the MINERVE reactor [3] to qualify this nuclear data. The interest of this technique is to provide an accuracy of 5% for radionuclides which are very poorly characterised because their γ-ray spectra have been incompletely measured or because the source preparation cannot be done in a reasonable time compared to the radionuclide half-life.

In this work, new values for the most intense γ-ray emission probabilities are proposed for ^{171}Er, ^{181}Hf, ^{233}Pa and ^{243}Pu. After a brief description of the pile oscillation and activation techniques, the experimental results are presented and compared to the JEFF3.1 [4] evaluated values.

2 Description of the experimental techniques

2.1 The MINERVE reactor

MINERVE is a pool type reactor operating at a maximum power of 100 W. MINERVE is dedicated to neutronics studies of different reactor types by means of high-precision measurements of the reactivity effects of small natural UO_2 fuel samples, doped with separated isotopes. The MINERVE core is composed of a driver zone, consisting of HEU plate-type MTR elements and surrounded by a graphite reflector, and a central cavity which houses the experimental zone with a lattice of fuel rods. For the experiments presented below, a UO_2 PWR type spectrum is considered and consists of UO_2 rods with an enrichment in ^{235}U of 3%.

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2.2 Reactivity measurements

The oscillation technique [5] consists in placing alternatively different samples in the center of the core in order to measure the associated reactivity variation. The center of the lattice is occupied by a rod-like oscillation device, which permits to place alternatively two different samples in the mid-plane of the reactor. One of these samples is the reference one, placed in the bottom of the oscillation rod, while the other with the material to be measured is charged in the upper part.

During the measurement, the rod moves periodically between an upper and a lower position. A Boron chamber, placed in the driver zone, detects the small variation of the reactor power induced by the oscillation of the samples and commands a rotary absorber in order to compensate the reactivity variation and to maintain criticality.

The reactivity effect is measured in terms of absorber rotation angle (or "pilot units") and is therefore not directly comparable with the results of the neutronics calculations (expressed in pcm or "calculation units").

The normalization is achieved by measuring a series of seven very well characterized UO_2 samples with enrichments in ^{235}U from 0.25% to 4.9%. They represent a very precise variation of a single neutronics parameter, which is supposed to be calculated perfectly. Figure 1 plots the measured reactivity effect (ordinate) against the calculated reactivity effect (abscissa). The slope of the regression line gives the factor to convert the pilot units to calculation units.

The interpretation is performed using the French deterministic transport code APOLLO2 [6]. Reactivity worth is obtained using the exact perturbation theory:

$$\delta\rho = -\frac{\langle\phi^*, \delta H\phi'\rangle}{\langle\phi^*, F'\phi'\rangle}. \tag{1}$$

The APOLLO2 code is a modular code, which solves both the Boltzmann integral equation and the integro-differential equation. The recent APOLLO2 version uses two nuclear data libraries, which are processed from the European previous JEF-2.2 and new JEFF-3.1 files. Two multigroup structures are available: the XMAS-172g and the new SHEM-281g meshing [7] (which avoids mutual and self-shielding treatments below 22 eV).

The analysis of our experiment allows a limited multicell geometry [8]: a 2D 11 × 11 cells pattern is used (see fig. 2). The pattern is a regular UO_2 lattice including the oscillation tube and sample in the central cell.

The space-dependent self-shielding calculation (4 rings in every fuel pin) for resonant isotopes is performed using the Probability Table method. APOLLO2 calculation uses P_{ij}-2D-UP1 model (interface current method with linear anisotropic angular fluxes).

The difference between an admixed sample signal ($^{Nat}UO_2$ + isotope X) and the $^{Nat}UO_2$ sample signal corresponds to the isotope reactivity worth. The experimental validation of this difference provides trends on the X(n,abs) cross section:

$$(C/E)_{oscillation} = \frac{X(n, abs)|_{calc}}{X(n, abs)|_{\exp}}. \tag{2}$$

The overall uncertainty includes the experimental uncertainty due to the signal difference (~1%), the uncertainty due to the material balance (~1%, mainly due to the isotope content) and the uncertainty due to the calibration (1.3%).

2.3 Activation measurements

In complement with the pile oscillation experiments, a post-irradiation technique [9] for measuring specific reaction rates, is applied on the samples irradiated in the MINERVE reactor. These experiments are dedicated to the qualification of neutronics power distribution inside the core or to the qualification of integral cross section for specific isotopes.

The measurements consist of a γ-ray spectrometry analysis of the irradiated fuel samples, in order to determine characteristic capture or fission products, linked to the corresponding reaction rates. The experimental device consists of a LEGe detector with a high resolution at low energies, a CANBERRA DSP2060 for signal processing and an AIM Multi-channel Analyser which provides data storage and analysis via the CANBERRA GENIE2000 software. The feasibility of the experiment depends on some properties of the reaction product, like its radioactive period, the energy and emission probability of the studied γ-ray.

Resolving the evolution equations during the different phases of the experiment – irradiation, cooling and counting –, the (n,γ) capture rate on the X isotope is obtained as follows

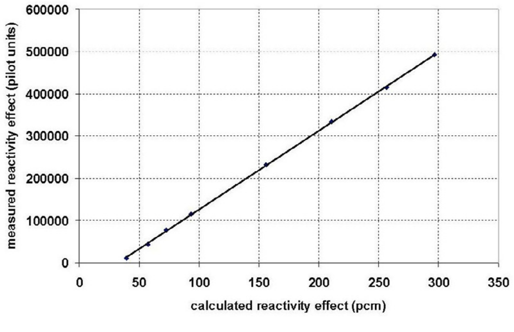

Fig. 1. Calibration of the absorber rotation angle with regard to the calculated reactivity effect. Error bars are given at 2σ of measurement uncertainty.

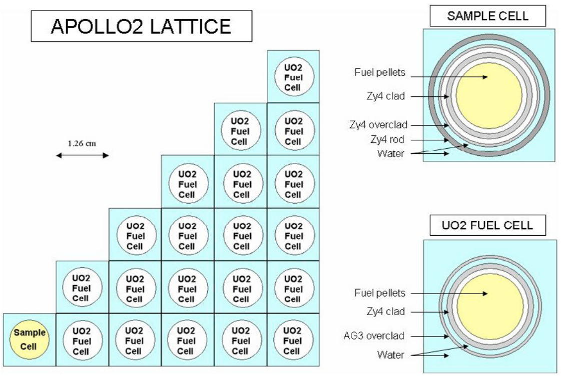

Fig. 2. APOLLO2 calculation pattern for the interpretation of the oscillation experiments.

[9]:

$$X(n,\gamma) = \frac{n_{sat}(E)}{I(E)R^P(E)T(E)} \tag{3}$$

$n_{sat}(E)$ is the saturated count rate of the γ-ray with the energy E, $I(E)$ the emission probability of the γ-ray with the energy E emitted after the decay of the studied reaction product, $R^P(E)$ the full absorption efficiency for a point source with the energy E and $T(E)$ the efficiency transfer correction.

The saturated count rate $n_{sat}(E)$ is obtained from net peak area measurements using the following relation:

$$n_{sat}(E) = \frac{N(E)}{t_r} C_\theta C_{dec} \tag{4}$$

$N(E)$ is the net area of the photopeak with the energy E, t_r the real time of measurement, C_θ and C_{dec} are the dead time and decay correction factors.

To avoid the measurement of the neutron flux inside the sample during the irradiation, the capture rate on X is always normalized to the reaction rate of a reference isotope. In our experiments, ^{238}U is considered for the accuracy of its (n,γ) cross section and of the γ-ray emission probabilities of the capture product (^{239}Np). As a consequence, the ratio of these reaction rates can be written as follows:

$$\frac{X(n,\gamma)}{^{238}U(n,\gamma)} = \frac{n_{sat,X}(E)}{n_{sat,^{238}U}(E_o)} \frac{I_{^{239}Np}(E_o)}{I_X(E)} \frac{R^P(E_o)}{R^P(E)} \frac{T(E_o)}{T(E)}. \tag{5}$$

For our experiments, $E_o = 277.6\,\text{keV}$ is the reference γ-ray with the better accuracy in the associated nuclear data.

For the interpretation of the activation experiments, the same calculation scheme as described in the previous section is used. The comparison between the calculation and the experiment provides trends on the X(n,γ) cross section :

$$(C/E)_{activation} = \frac{X(n,\gamma)|_{calc}}{X(n,\gamma)|_{\exp}} \frac{^{238}U(n,\gamma)\big|_{\exp}}{^{238}U(n,\gamma)\big|_{calc}}. \tag{6}$$

2.4 Emission probability measurements

For non fissionable isotopes or if the fission part of the cross section is low enough compared to the (n,γ) cross section, the two experimental techniques bring complementary information on the X(n,γ) cross section under the assumption that the decay data used in the activation measurements are correctly evaluated. Consequently, any significant difference between the C/E ratio obtained for the oscillation technique and the C/E ratio obtained for the activation technique can be correlated to an error on the γ-ray emission probability of the capture product. This can be written as follows:

$$I_X(E) = I_{^{239}Np}(E_o) \frac{^{238}U(n,\gamma)}{X(n,\gamma)} \frac{n_{sat,X}(E)}{n_{sat,^{238}U}(E_o)} \frac{R^P(E_o)}{R^P(E)} \frac{T(E_o)}{T(E)}. \tag{7}$$

The ratio of capture rate is estimated as follows:

$$\frac{^{238}U(n,\gamma)}{X(n,\gamma)} = \frac{^{238}U(n,\gamma)}{X(n,\gamma)}\bigg|_{calc} \frac{(C/E)_X}{(C/E)_{^{238}U}}. \tag{8}$$

The overall uncertainty on the γ-ray emission probability $I_X(E)$ is calculated from the variance propagation relation:

$$\left(\frac{u(I_X(E))}{I_X(E)}\right)^2 = \left[\begin{array}{c} \left(\frac{u(I_{^{239}Np}(E_o))}{I_{^{239}Np}(E_o)}\right)^2 + \left(\frac{u((C/E)_X)}{(C/E)_X}\right)^2 + \\ \left(\frac{u((C/E)_{^{238}U})}{(C/E)_{^{238}U}}\right)^2 + \left(\frac{u\left(\frac{n_{sat,X}(E)}{n_{sat,^{238}U}(E_o)}\right)}{\frac{n_{sat,X}(E)}{n_{sat,^{238}U}(E_o)}}\right)^2 + \\ \left(\frac{u\left(\frac{R^P(E_o)}{R^P(E)}\right)}{\frac{R^P(E_o)}{R^P(E)}}\right)^2 + \left(\frac{u\left(\frac{T(E_o)}{T(E)}\right)}{\frac{T(E_o)}{T(E)}}\right)^2 \end{array}\right]. \tag{9}$$

3 Experimental results

Our study focuses on the emission probability of the most intense γ-ray emitted by the three following isotopes [5][8]: ^{171}Er, ^{181}Hf and ^{243}Pu, respectively produced by (n,γ) reactions on ^{170}Er, ^{180}Hf and ^{242}Pu. To check the consistency of our approach, we also have measured the emission probability of the 312.0 keV γ-ray of ^{233}Pa, produced by (n,γ) capture on ^{232}Th, to compare to the JEFF3.1 evaluation, which is supposed to be known with a high precision.

The reference γ-ray emission probability is taken from JEFF3.1 library:

$$I_{^{239}Np}(277.6\,\text{keV}) = (14.38 \pm 0.43)\%.$$

The ratio of the saturated count rates and the ratio of the full absorption efficiencies are deduced from the experiment. The ratio of the efficiency transfer corrections are calculated from Monte Carlo calculations, simulating the full three dimension geometry of the experimental device and estimating the self-absorption inside the fuel sample for the energies E and E_o.

The $(C/E)_X$ ratio can be assimilated to the C/E ratio for the oscillation experiments. In the case of ^{238}U, the qualification studies of the APOLLO2 code have shown that the (C/E) ratio for the ^{238}U capture rate is (1.017 ± 0.020) [6].

Table 1 gives the experimental results obtained, combining oscillation and activation measurements with the uncertainty budget detailed in table 2.

4 Discussion

Table 1 shows significant discrepancies between the JEFF3.1 evaluations and the measured value, for ^{171}Er [10], ^{181}Hf [11]

Table 1. Experimental results of γ-ray emission probability measurements (uncertainties are given at 1σ).

Isotope	E (keV)	**Measured $I(E)$ ($\times 10^{-2}$)**	JEFF3.1 $I(E)$ ($\times 10^{-2}$)	(J3.1 – M) / M
^{171}Er	308.3	**50.1 ± 7.7**	64.4 ± 2.5	(19.1 ± 20.5)%
^{181}Hf	482.2	**67.7 ± 7.2**	80.6 ± 0.2	(28.6 ± 12.7)%
^{243}Pu	84.0	**19.2 ± 1.0**	23.0 ± 2.0	(20.0 ± 12.3)%
^{233}Pa	312.0	**40.3 ± 1.8**	38.5 ± 0.4	(−4.4 ± 4.7)%

Table 2. Uncertainty budget on the γ-ray emission probability measurements (uncertainties are given at 1σ).

Parameter	^{171}Er	^{181}Hf	^{243}Pu	^{233}Pa
$I_{^{239}Np}(E_o)$	3.0%			
$(C/E)_{^{238}U}$	1.5%			
$(C/E)_X$	15%	10%	3.0%	3.0%
$\frac{n_{sat,X}(E)}{n_{sat,^{238}U}(E_o)}$	0.39%	0.40%	2.5%	0.52%
$\frac{R^P(E_o)}{R^P(E)}$	0.10%	0.50%	0.60%	0.15%
$\frac{T(E_o)}{T(E)}$	0.30%	0.30%	0.50%	0.30%
Total relative uncertainty	**15.4%**	**10.6%**	**5.22%**	**4.54%**

and ^{243}Pu [12]. For the two first ones, uncertainties on the reactivity measurement (i.e., on the $(C/E)_X$ ratio) have been deliberately overestimated due to an experimental problem during the calibration phase. New measurements are scheduled in mid 2007 with target uncertainties below 5%.

Nevertheless, these preliminary results enlighten a potential error on the JEFF3.1 evaluations, especially for ^{181}Hf which deviates by 29% with our measured value, corresponding to more than 2σ of uncertainty.

For ^{243}Pu, a discrepancy of 20% is observed with the JEFF3.1 value. A reduction by a factor 2 is achieved with our technique, compared to the reference studies.

The overestimation of the JEFF3.1 values with respect to our measurements for these three isotopes may be partly due to the pandemonium effect, as the missing of energy levels may involve an underestimation of the normalisation factor in the absolute calibration of γ-ray emission probabilities.

For the ^{233}Pa, whose normalization factor in the γ-ray emission probability is the combination of seven independent measurements, a consistency better than 1σ of uncertainty is observed with the JEFF3.1 value [13]. This result tends to validate our technique for a supposed well known isotope.

5 Conclusion

In this paper, an innovative method, combining oscillation and activation experiments, has been presented to measure the emission probabilities of some γ-rays which could not be easily measured with usual techniques. The target uncertainty of the method, around 5%, involves a significant improvement in the knowledge of rare isotopes. The technique has been applied to ^{171}Er, ^{181}Hf, ^{243}Pu and ^{233}Pa:

- For the three first ones, an overestimation of 19% to 29% of the JEFF3.1 evaluated emission probability of the most intense γ-ray has been identified and may be partly due to the pandemonium effect. The current uncertainties for ^{171}Er and ^{181}Hf, respectively 15% and 10%, will be significantly reduced in the future with a new measurement campaign and a target uncertainty below 5%.
- For ^{233}Pa which is supposed to be well estimated in the nuclear data libraries, an accordance within 1σ of experimental uncertainty is observed between our measurement and the JEFF3.1 evaluation.

Further studies will be undertaken in 2008 with the same technique, on the following isotopes: ^{238}Np, ^{244}Am.

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