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### SINRD validation experiments at the time-of-flight facility GELINA



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#### ABSTRACT

Self-interrogation neutron resonance densitometry (SINRD) is a non-destructive analysis technique that can be used to quantify the amount of <sup>239</sup>Pu in spent nuclear fuel. It is a passive method that relies on the detection of neutrons, which are emitted by the fuel. The amount of <sup>239</sup>Pu is estimated from the ratio of the neutron intensity in the fast energy region and in a region close to the 0.296 eV resonance of <sup>239</sup>Pu. The neutron intensity in the resonance region is obtained from a detection system with a high sensitivity to 0.296 eV neutrons. This can be realized by using two neutron detectors with <sup>239</sup>Pu as convertor material. One of the detectors is covered by a thin Gd foil and the other by a thin Cd foil. The Gd and Cd foils are referred to as SINRD filters.

An approach based on the measurement of a fuel assembly in air and surrounded by a slab of polyethylene was developed at SCK-CEN. This approach foresees the insertion of small neutron detectors in the guide tubes of the assembly, and optimisation studies of SINRD were based on Monte Carlo simulations. Experiments to support the results of such simulations were carried out at the time-of-flight facility GELINA of the Joint Research Centre (JRC) in Geel (Belgium). Transmission measurements were performed to verify the quality of the nuclear data that are used to define the optimum thickness of the SINRD filters. Results of self-indication measurements were used to confirm the basic principle of SINRD, that is, that the best results are obtained with a detector that has a high sensitivity to neutrons with energy close to the energy of a strong resonance of the material under investigation. The results of these experiments are presented in this work.

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#### 1. Basic principles of SINRD

Self-Interrogation Neutron Resonance Densitometry (SINRD) (LaFleur, 2011; LaFleur et al., 2012a,b; Hu et al., 2012; Hu et al., 2013; LaFleur et al., 2013; LaFleur et al., 2015) is a passive neutron technique to determine the amount of fissile material in spent fuel. It originates from a technique proposed in 1968 (Menlove et al., 1969) which was based on active interrogation with an external neutron source. This technique, referred to as Self-Indication Neutron Resonance Absorption Densitometry, was inspired by the basic principles of self-indication measurements. The latter is a well-known technique to study cross section data in the resonance region (Fröhner et al., 1966). Applying SINRD to spent fuel, the interrogation relies on prompt fission neutrons from spontaneous

fission of <sup>244</sup>Cm present in the fuel without the use of an external neutron source. Therefore, the term self-interrogation was introduced by LaFleur (2011).

The total microscopic cross-section for  $^{239}$ Pu is reported in Fig. 1 for neutron energy close to the resonance at 0.296 eV. In addition, the transmitted neutron fluxes calculated in air through two homogeneous samples containing  $^{238}$ U and different percentages of  $^{239}$ Pu are shown. The samples had a density of  $10.4 \text{ g/cm}^3$  and thickness equal to the radius of a fuel pin in a PWR  $17 \times 17$  fuel assembly, i.e. 0.4025 mm. The figure shows the corresponding attenuation of the neutron flux at the energy of the  $^{239}$ Pu resonance, and indicates that the attenuation is more evident for the sample with higher quantity of  $^{239}$ Pu. The attenuation of the neutron flux observed in Fig. 1 can be related to the  $^{239}$ Pu content in the spent fuel. Such attenuation is measured with SINRD by calculating the SINRD signature ( $R_{SI}$ ) according to Formula (1).

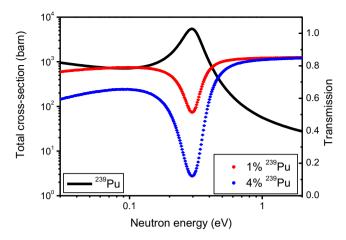
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**Fig. 1.** Total cross-section of <sup>239</sup>Pu (left axis) and transmitted neutron fluxes through <sup>239</sup>Pu samples (right axis) as a function of the neutron energy.

$$R_{SI} = \frac{C_F}{C_{Gd} - C_{Cd}} \tag{1}$$

The SINRD signature was defined in (LaFleur, 2011) as the ratio between the neutron intensity in the fast energy region and in a region close to the <sup>239</sup>Pu resonance.

The use of SINRD for spent fuel measurement was first proposed by (LaFleur, 2011) by measuring spent fuel under water, and by placing a set of <sup>235</sup>U fission chambers on one side of the fuel assembly. This differs significantly from the SINRD approach proposed by SCK•CEN (Rossa et al., 2014; Rossa et al., 2015a,b) that focuses on measurements of PWR spent fuel assemblies in dry conditions. Neutrons emitted from the fuel are measured with small detectors which are inserted in the guide tubes of a nuclear fuel assembly. According to our approach (Rossa et al., 2015a), the neutron intensity in the fast region is derived from the response of a <sup>238</sup>U fission chamber  $(C_F)$ , while the neutron intensity in the resonance region is taken as the difference between the neutron counts of two <sup>239</sup>Pu fission chambers covered with Gd and Cd foils, denoted by  $C_{Gd}$  and  $C_{Cd}$ , respectively. The choice a <sup>239</sup>Pu fission chambers allows the implementation of the self-indication principle (Fröhner et al., 1966), with an increased sensitivity to the neutron flux around the energy region close to the <sup>239</sup>Pu resonance. According to the self-indication principle, one utilizes the same sample material, <sup>239</sup>Pu in this case, both in the detector and in the sample attenuating the flux being measured.

#### 2. Validation experiments at GELINA

Most of the optimization studies of SINRD, e.g. (LaFleur et al., 2012a; Hu et al., 2013; Rossa et al., 2015a,b) are based on results of Monte Carlo simulations. The optimum characteristics of the SINRD filters were defined by Rossa et al. (2015b) based on theoretical calculations. These characteristics differ from those proposed by LaFleur et al. (2012a). To validate the results of Rossa et al. (2015b), transmission experiments were carried out at the time-of-flight (TOF) facility GELINA. A detailed description of this facility, which is operated by the Joint Research Centre, Geel, Belgium, can be found in Mondelaers et al. (2006).

Transmission factors through well-characterized Cd and Gd samples of different thickness were obtained and compared with the obtained ones when using the data libraries used for the optimization (Rossa et al., 2015b). In addition, self-indication experiments were carried out to demonstrate the basic principles of SINRD, and an increased sensitivity to the neutron flux around a given energy region can be achieved by utilizing the same sample

material both in the detector and in the sample attenuating the flux.

#### 2.1. Setup of the transmission experiments

Transmission measurements can be used for the determination of total neutron cross-section data (Fröhner et al., 1966; Massimi et al., 2011; Schillebeeckx et al., 2014), and the schematic view of the measurement setup is shown in Fig. 2. The experiments were carried out to measure the transmitted neutron flux ( $\phi_{tr}$ ) through several samples of Gd and Cd as a function of the time-of-flight.

Transmission measurements were carried out at a 10 m transmission station of GELINA (Paradela et al., 2015), with the accelerator operating at 50 Hz. The moderated neutron spectra originated from GELINA can be approximated by a Maxwellian distribution in the thermal region, a  $1/E^{0.85}$  dependence in the epithermal region, and a Watt function in the fast region (Schillebeeckx et al., 2014). A detailed view of the experimental setup is shown in Fig. 3. An automatic sample changer is positioned at 7.7 m from the neutron producing target, allowing an automated alternation of sample-in and sample-out measurements. A second sample changer, which is placed close to the sample position, is used for anti-overlap and background filters. Neutrons passing through the sample and the filters are detected by a 6.35 mm  $\times$  76 mm  $\times$  76 mm Li-glass scintillator, placed at a 11 m distance from the neutron producing target. The detector, which is enriched to 95% in <sup>6</sup>Li, is directly viewed by one photomultiplier tube. A good transmission geometry is realized by proper collimation of the neutron beam. A set of Li, B, Cu, Ni and Pb collimators with decreasing diameter are placed between the neutron producing target and the sample; a similar sequence of collimators with increasing diameter are placed between the sample and the detector. Experiments in good transmission geometry are required to assure that all detected neutrons traverse the sample and scattered neutrons do not reach the detector (Schillebeeckx et al., 2012).

Transmission measurements using Gd and Cd metal foils or discs of different thicknesses were carried out. The characteristics of the samples are summarized in Table 1. The beam at the sample position had a 10 mm diameter. To reduce the influence of the  $\gamma$ -ray flash in the detector a permanent Pb filter was installed in the beam line.

The experimental transmission,  $T_{\text{exp}}$ , was obtained from the ratio of a sample-in measurement  $C_{\text{tr}}$  and a sample-out measurement  $C_0$ , both corrected for their background contributions  $B_{\text{tr}}$  and  $B_0$ , respectively:

$$T_{\rm exp} = \frac{C_{tr} - B_0}{C_{tr} - B_0} \tag{2}$$

The TOF spectra ( $C_{\rm tr}$ ,  $B_{\rm tr}$ ,  $C_0$ ,  $B_0$ ) in Eq. (2) were corrected for losses due to the dead time in the detector and electronics chain, and all spectra were normalized to the same neutron intensity and TOF-bin width. The background, which is a sum of TOF

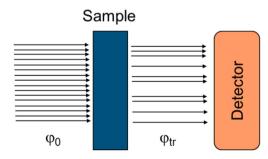


Fig. 2. Schematic view of a transmission experiment.

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