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Improvement of the activity measurement method for solid dosimeters emitting X-rays

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HIGHLIGHTS

- Measurements of X-rays require significant corrections for self-attenuation and fluorescence.
- Provide experimental validation for self-attenuation corrections.
- Improvement of uncertainty of emission intensities for ^{93m}Nb and ^{103m}Rh.
- Validation of this parameters for characterize the neutron flux around 1 MeV more precisely.

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ABSTRACT

Today, there is growing interest for neutrons in the intermediate energy range between 100 keV and 1 MeV, which are responsible for damaging materials in reactor. To improve this deficiency, we use rhodium and niobium which, through the inelastic neutron scattering reaction, leads to the formation of ^{103m}Rh and ^{93m}Nb low-energy X-emitters. This paper describes the improvements and validation made on this type of complex measurement by X spectrometry: self-attenuation, fluorescence correction, and emission intensity were poorly known previously.

1. Introduction

The "Laboratoire de Dosimétrie, Capteurs et Instrumentation" (LDCI) located at the CEA Cadarache center (France) is in charge of measuring the activity of samples (or dosimeters) placed in irradiation reactors, power reactors and critical mock-ups. The type of sample used is selected on the basis of its energy sensitivity to incident neutrons. For many years, physicists focused on low-energy neutrons, called thermal neutrons, to gain a better understanding of the power of the different reactors. The neutron energy range above 1 MeV then attracted the interest of material specialists who assumed that fast neutrons were the main cause of damage to materials. Today, there is growing interest in neutrons in the intermediate energy range between 100 keV and 1 MeV, which are also responsible for damaging materials. Nonetheless, there is still sustained interest in the energy region around 1 MeV and above. In fact, there is a continued, if not increasing demand for a more in-depth knowledge of material damage. However, the computational codes and the nuclear data libraries commonly used still need to be enhanced.

Niobium and rhodium are two materials used as solid dosimeters to meet the needs described above. Inelastic scattering reactions (n,n′) lead to the formation of ^{93m}Nb and ^{103m}Rh, X-ray emitting radionuclides which provide the data required, through activity measurement (expressed in Bq), on this region of the fast neutron spectrum. Niobium is the most relevant dosimeter for monitoring damage to pressurised reactor vessels due to its response threshold (around 1.2 MeV) and its long half-life (16.12 years) over long periods of exposure to neutron flux. Rhodium, with its short half-life (56 min), is reserved for use as a dosimeter in critical mock-ups to characterise neutron flux above 700 keV.

X-ray photons are emitted by niobium and rhodium in low energy ranges. Measurements of these X-rays require significant corrections for self-attenuation and fluorescence associated not only with the dosimeter material itself but also with impurities. They are also characterised by high uncertainties due to the lack of knowledge of their

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emission intensities.

The purpose of this study is to present the recent improvements on these factors which adversely affect measurements (nuclear data, fluorescence correction), and to provide experimental validation for self-attenuation corrections. These advances will allow us to apply the refined activity measurements to neutron computations in order to characterise this spectral region more precisely. The set of results presented in this paper is expressed with standard combined uncertainties ($k = 1$).

2. Limitations of X-ray measurements at the madere platform

The MADERE reactor dosimetry activity measurement platform (Girard et al., 2009) is part of the LDCI lobratory is used to measure the X-ray activity of solid niobium and rhodium dosimeters with thicknesses of 20 μm and 50 μm respectively. It uses two measurement techniques:

- Activity measurement by a relative method: X-ray spectrometry through direct comparison of a reference sample with a measurement sample. This relative measurement technique is only used for ^{93m}Nb. The short radioactive half-life of ^{103m}Rh makes it impossible to maintain a reference sample. It is a highly controlled, relatively simple technique that involves comparing two counting rates. This type of measurement requires to apply a gamma induced fluorescence correction on niobium measured data. The evaluation of this correction dates back to 1973 (Lloret, 1973). Elsewhere, the relative uncertainties on the emission intensities of the K X-ray lines for ^{93m}Nb are around 2%. The squality of the knowledge of both the fluorescence correction and the emission intensities will have a direct impact on the reference sample activity measurement.
- Activity measurement using efficiency calibration curve: This measurement technique can be used for niobium and rhodium. Traditionally used to measure gamma rays, this is a more complex method for measuring low-energy X-rays. Besides an exact knowledge of the measurement environment (Domergue et al., 2009), it requires controlling the characteristics of the measurement sample and the resulting correction factors. This method also poses problems relating to fluorescence corrections and the relative uncertainties of the emission intensities of ^{93m}Nb and K X-ray lines of ^{103m}Rh. These are currently between 5.5% and 7%. This method also involves self-attenuation corrections of between 20% and 50% depending on the thickness of the dosimeter, which must be controlled given their impact on the final result. An initial approach was conducted in 2009 (Domergue et al., 2009); this study will allow this correction to be validated experimentally.

Progress on the various parameters presented in this study will help to improve the measurement uncertainties for ^{93m}Nb and ^{103m}Rh: a target of 1.5% relative combined standard uncertainty associated with activity is sought. This will allow us to make full use of activity measurements to refine the computational codes for predicting reactor physics in an energy region around 1 MeV.

3. X-ray activity measurement at the madere platform

The MADERE platform has gained testing accreditation from the French certification body COFRAC for mass activity measurements by gamma and X-ray spectrometry of solid samples irradiated. This measurement platform plays a supporting role in an approach to validate computational codes, monitor the degradation of materials subjected to intense fast neutron flux, conduct experimental programmes and take part in reactor start-up tests.

For the purposes of X-ray spectrometry, the MADERE platform is equipped with 2 measuring channels (X1 and ×2). These are instrumented with LEGe detectors equipped with a thin beryllium window. We use digital electronics and CANBERRA data acquisition software. Measured spectra are processed using COLEGRAM software (Ruellan et al., 1996). The measurement environment is optimised as follows in an attempt to minimise scattering phenomena which disturb X-ray spectra. Samples are positioned using a device made from the smallest possible amount of a light material (plexiglass) and collimation uses two tantalum windows.

3.1. Activity measurement by relative method

Only this type of measurement is applied nowadays for the detection of ^{93m}Nb produced in niobium dosimeters.

The activity measurement involves 3 stages:

- In France, the Laboratoire National Henri Becquerel (LNHB) - the French metrology laboratory for ionising radiations - supplies a calibration certificate for a single reference standard (radionuclide / activity).
- The MADERE platform performs an "internal connection" of working standards, covering a wide range of activity, to this reference standard.
- The MADERE platform takes activity measurements of dosimeters relative to the working standards. A working standard is selected based on its degree of similarity of activity to the dosimeter in question.

Relative measurements impose the use of dosimeters of the same dimensions as those used in the standards (working and reference) consisting exclusively of 20 μm thick Nb strips.

The X-ray specific activity (A_i) measurement of the sample studied is given in relation to the working sample based on the following formula:

$$A_i = \frac{\mathscr{A}_{work\ j}}{m_i} \cdot \frac{\tau_i}{\tau_{work\ j}} \cdot C_{fluo\ i} \cdot C_{t0} \quad (1)$$

$A_{(work\ j)}$Activity of the working standard j (Bq)
m_iMass of the dosimeter i (mg)
τ_iAverage emission rate of the sample i (s^{-1})
$\tau_{(work\ j)}$Average emission rate of the working standard j (s^{-1})
$C_{(fluo\ i)}$Fluorescence correction of the sample i
$C_{(t0)}$Correction to return the activity of the working standard to the required date

Given the high radioactive half-life of niobium (16.12 years) compared with the acquisition time (maximum of 7 days), activity decay during the sample measurement time (working standard or dosimeter) is negligible (far less than 0.1%) and thus disregarded in (Eq. (1)).

The fluorescence correction described in IV.1.3 is poorly controlled (the last re-assessment of fluorescence coefficients dates back to 1973 (Lloret, 1973). This is also true of the relative standard uncertainties of the emission intensities of the ^{93m}Nb 16.5 keV and 18.6 keV lines, which are around 2% and could be improved (Bé et al., 2016). These uncertainties have a direct impact on the activity value of the working standard, which is itself impacted by the activity value of the LNHB's reference standard (3.2%).

3.2. Activity measurement using an efficiency calibration curve

This type of measurement is applied for the detection of ^{103m}Rh (Domergue et al., 2009), (resp. $^{93\ m}$Nb) produced in rhodium (resp. niobium) irradiated dosimeters.

The rhodium used is in the form of 8 mm diameter disks in thicknesses of 6, 12 and 50 μm, respectively. These dimensions are not restrictive, but correspond to the stock of dosimeters available for the MADERE platform. The niobium dosimeters have the same dimensions as those detailed in section III.1.