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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.

X-ray activity measurements based on an efficiency calibration curve differ from conventional measurements for activation dosimeters for two reasons:

- Rhodium and niobium dosimeters are extremely thin in order to minimise self-attenuation corrections and need to be kept between two sheets of Mylar during the measurement. The standard sources are also encased between two Mylar sheets but with thicknesses different from the dosimeter ones. A correction is therefore necessary to take account of the proportion of X-rays absorbed by these different thicknesses.

- K_α and K_β X-ray lines are both used to determine the dosimeters activity. This arrangement is necessary due to the level of knowledge regarding emission probabilities, which is poor for ^{103m}Rh and could be better for ^{93m}Nb. The use of these two lines significantly reduces the uncertainty of the final result. Each of these lines is the composed of several "sub lines" $K_{\alpha1}$ and $K_{\alpha2}$, or $K_{\beta1}$, $K_{\beta2}$, etc…., which are deconvoluted.

The dosimeter specific activity A_i, measured from emission of an X-ray peak, is given by the following formula:

$$A_i = \frac{S_i}{m_i . \varepsilon_{dét} . I_X} . C_{s.a} . C_{Fluo} . \prod_i C_i \quad (2)$$

m_iMass of the dosimeter i (mg)
S_iCounting rate under full-energy peak with energy "E_i"(s^{-1})
ε_{det}Detection efficiency at energy "E_i"
I_γEmission intensity of the X-ray line with energy "E_i"(s^{-1})
ε_{det}Detection efficiency at energy "E_i"
$C_{s.a}$Self-attenuation correction of X-rays in the dosimeter
C_iCorrection factors (decay during acquisition, coincidences, return to the required date,…)
C_{Fluo}Correction coefficients for fluorescence

As is the case for relative measurements, knowledge of fluorescence corrections and the relative uncertainties of ^{93m}Nb and ^{103m}Rh emission intensities (about 2% and 5–7% respectively) is poor.

Self-attenuation corrections between 20% and 50% depending on the dosimeter can affect the precision of the expected end results by around 1.5%. In this study, the self-attenuation factor were determined then validated by means of two separate experiments:

- Comparison of the activity of a 20-µm thick solid niobium strip (width: 1 mm and length: 5 mm) measured at MADERE with the activity measured after dissolution (Song et al., 2006) at the CEA LARC laboratory.
- Comparison of the mass activity measurement of 3 rhodium disc (diameter: 8 mm) of different thicknesses (6, 12 and 50 µm) irradiated in EOLE, experimental mock-up reactor (Thiollay et al., 2014)

4. Data improvements program

4.1. Refining the modelling process

4.1.1. Modelling the efficiency calibration curve

The efficiency calibration of ×1 and ×2 is defined for a reference geometry using standard radioactive point sources supplied by "Compagnie pour l'Etude et la Réalisation de Combustibles Atomiques / Laboratoire d'Etalons d'Activité (CERCA/LEA)": Y-88, Ba-133, Cs-137, Co-57, Sn-113 m, Ce-139, Ba-133 and Am-241. Three efficiency calibration curves are obtained for different distances between the detector and the point source:

X1−E4 = 42 mm.

X2−E4 = 43 mm

X2−E8 = 93 mm.

The efficiency calibration ε(E) is derived according to:

$$\varepsilon(E) = \frac{N(E)}{A\, I(E)\, t} \prod_i C_i \quad (3)$$

$N(E)$Counting rate under full-energy peak with energy "E" (s^{-1})
AActivity of the radionuclide included in the standard point source (Bq)
$I(E)$Emission intensity of line with energy "E"
tSpectrum measurement time (live time in seconds)
C_iCorrection factors

The experimental values are adjusted using the least-square method based on a polylogarithmic function (Morel, 1996) to obtain the best fit for an efficiency value on the energy range between 10 keV and 140 keV (approximately). The experimental efficiency values are obtained with 1.5% relative combined standard uncertainties.

Due to the scant knowledge of photon emission intensities in the low energy range, the efficiency calibration curve was refined using the Monte Carlo simulation method. The PENELOPE (Salvat et al., 2015) code was used and the physical parameters of the germanium crystal were optimised to adjust the efficiency curve obtained by simulation to the experimental fit. The geometry of the germanium detector was established based on the dimensions supplied by the manufacturer and from the radiography of the detector (Fig. 1).

The detector efficiency was obtained using PENELOPE after several modifications of the crystal dimensions. Experimental (Eq. (3)) and simulation results for the ×2-E8 geometry are presented on Fig. 2. The relative differences between the experiments and the simulations are still between 2% and 4%. The modelling of the detector need to be refined, starting in particular with a more in-depth knowledge of germanium dead layers.

4.1.2. Modelling the self-attenuation correction

Three methods have been identified to determine the self-attenuation correction on rhodium and niobium samples (Lépy and Ménesguen, 2014) going from a simplified description of the case to a precise modelling:

- Simple analytical calculation based on material characteristics;
- Calculation of the efficiency transfer between a point source and the volume dosimeter with ETNA (Lépy et al., 2001);
- Monte Carlo simulation of the source-detector's full geometry with PENELOPE.

Fig. 3 shows the self-attenuation correction of the $X_{K\alpha}$ line (20.2 keV) for ^{103m}Rh for a 50 µm thick rhodium disc derived from the three methods by varying the distance between the source and the detector. Relative standard uncertainties are 3.0% (k=1).

Approximations on the description of the geometry differ widely depending on the method used. However, the results obtained are consistent within the associated uncertainties. At the same distance,

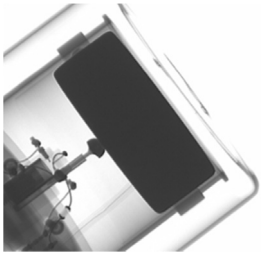

Fig. 1. Radiography of ×2 detector.

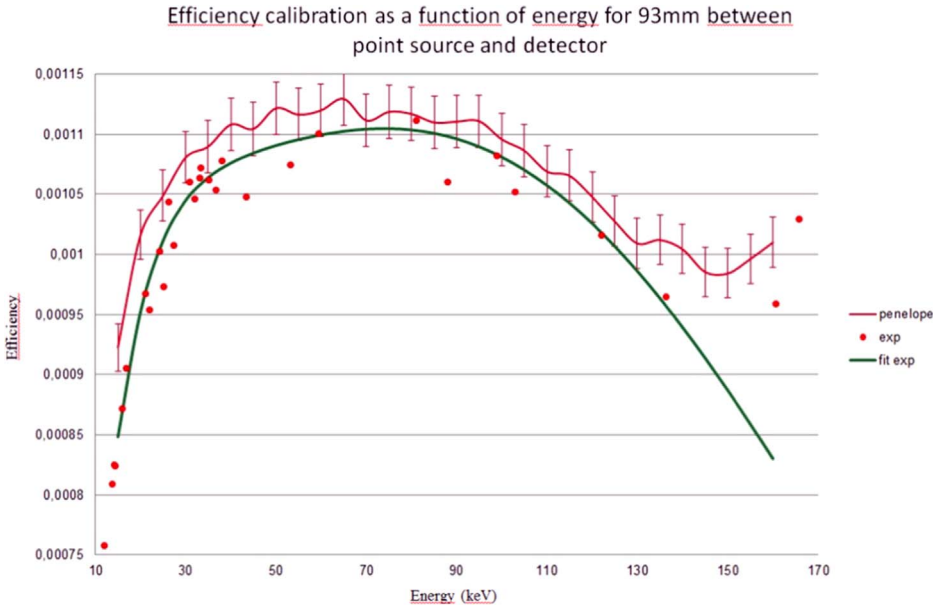

Fig. 2. Comparison between simulation and experimental efficiency of channel ×2-E8.

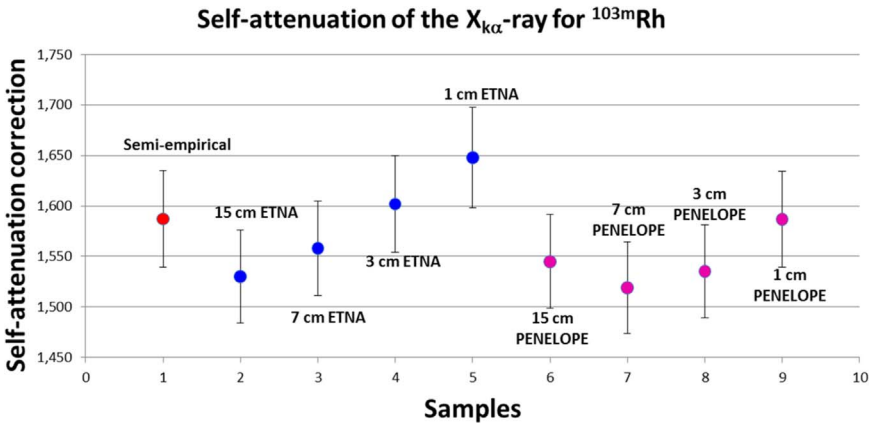

Fig. 3. Self-attenuation of the $X_{K\alpha}$-ray line of ^{103m}Rh ($k = 1$) in rhodium 50 μm thickness sample as a function of the measuring distance between sample and detector (cm).

results obtained using PENELOPE are 2–4% lower respectively than the approximated and ETNA values obtained.

On the other hand, the trend of the evolution of the self-attenuation correction as a function of the measurement distance is not consistent between ETNA and PENELOPE. This result needs to be further explored.

The analytical corrections are validated by the results obtained from ETNA and PENELOPE and can therefore be applied to correct the self-attenuation effect. If nuclear data on emission intensities with uncertainties around 1.5% are re-assessed, it will be necessary to review this validity.

4.1.3. Modelling the gamma induced fluorescence correction

This section focuses on the additional fluorescence induced in niobium dosimeters (Riffaud et al., 2016b). As already mentioned, the (n,n′) reaction on Nb-93 is sensitive to fast neutrons with E > 1 MeV. The neutron spectrum in a reactor also has a thermal and epithermal component triggering reactions with impurities. ^{182}Ta is thus activated by radiative capture from ^{181}Ta. Other radioactive isotopes of niobium are also created, such as ^{92m}Nb, ^{94}Nb by (n,2n) and (n, γ) reactions on ^{93}Nb. These radionuclides cause fluorescence phenomena (Fig. 4) which can disrupt the final $^{93\ m}$Nb activity result up to 10% according to the spectrum of the neutron flux in which the dosimeter is located.

When the dosimeter material is ionised, photoelectrons are ejected from niobium atoms. This phenomenon is followed by electron rearrangement, with emission of X-ray radiation. This fluorescence effect generates an extra emission of X-rays (red arrows in Fig. 4) which is added to that caused by the decay of ^{93m}Nb (green arrows in Fig. 4). Consequently, there is an overestimation of dosimeter activity and correction factors have to be taken into account to derive the activity of $^{93\ m}$Nb only. The presence of ^{60}Co impurities has also been observed occasionally in the past.

The ^{182}Ta and ^{60}Co depends on the impurity in the sample, while the ^{92m}Nb, ^{94}Nb and ^{95}Nb depends on the neutron flux.

The self-fluorescence coefficients used to date were calculated analytically in 1973 for ^{182}Ta, ^{94}Nb, ^{95}Nb and $^{92\ m}$Nb. The coefficients are expressed in s^{1}. Bq^{-1}, indicating the number of fluorescence X-rays emitted per second and per decay of the impurity (Lloret, 1973).

However, these coefficients were calculated only for the main

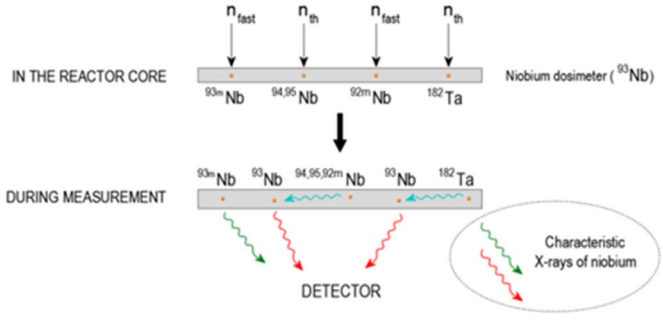

Fig. 4. Self-fluorescence in a niobium dosimeter.

Table 1
Fluorescence coefficients for impurities taken into account in activity measurements.

Impurity	Half- life	Values from 1973 (s^{-1}. Bq^{-1})	Values from 2017 (s^{-1}. Bq^{-1})	Relative difference $\frac{2017}{1973}-1$ (%)
^{92m}Nb	10.15 days	0.021	0.00003687 (296)	−99%
^{94}Nb	20,000 years	0.0055	0.0119 (2)	+116%
^{95}Nb	35 days	0.0033	0.00310 (6)	−6.1%
^{182}Ta	114.6 days	0.077	0.0645 (6)	−16.2%
^{60}Co	5.27 years	–	0.00821 (11)	–

energy lines and did not take account the electron (or positron) interactions for ^{182}Ta and ^{92m}Nb, or the self-attenuation of X-rays in niobium following electron rearrangement. The contributions of impurities to the typical dosimeter X-ray spectra were thus re-evaluated using the Monte Carlo method.

This work involved simulating the decay scheme for each radionuclide, taking into account the branching ratios, the electron and photon emission intensities and their interactions with the material and the atomic relaxation processes. The study (Riffaud et al., 2016b) gives a comprehensive description of the self-fluorescence coefficient calculation for each impurity based on spectra obtained through simulation for the niobium peaks.

Table 1 shows the results obtained, with standard uncertainties calculated according to the GUM (JCGM, 2008), and compares them with the coefficients evaluated in 1973.

The magnitude of corrections on dosimeter activity depends on the activity of each impurity. Fluorescence correction is negligible for ^{92m}Nb, especially since it has hardly ever been observed due to its short radioactive half-life. The ratios of fluorescence corrections observed between 1973 and this work are probably due to poor knowledge of the impact of emissions β in the first calculations.

^{94}Nb, in contrast, is present yet its activity is low due to its long radioactive half-life. Indeed, the fluorescence correction due to the presence of ^{94}Nb is less than 1%. The ratios of fluorescence corrections observed between 1973 and this work are due to the upgrading of the nuclear database (half-life and maximum energy of β spectrum).

Until now, the fluorescence correction due to ^{60}Co has not been taken into account. This is set to change. However, given the activities observed in ^{60}Co, this correction is declared to be minimal.

In practice, ^{182}Ta and are the most distorting radionuclides in terms of activity measurement.

It should be noted that the values in Table 2 were derived for a solid angle of 0.035 sr. The fluorescence corrections are only valid for a solid angle less than or equal to this. The study will be extended in the near future to include more similar measurement positions to that of the detector (solid angle of 0.15 sr) and to cover the full range of the MADERE platform's measurement positions. Lastly, the exercise will also be extended to include a significant quantity of β^+ emitting ^{185}W.

A similar study needs to be conducted for rhodium dosimeters to take account of iridium and platinum impurities.

4.2. Revision of ^{103m}Rh and $^{93\ m}$Nb nuclear data

Two experiments have been carried out to improve the emission intensities of $^{93\ m}$Nb and ^{103m}Rh:

- Determining the fluorescence yield of ^{93m}Nb and ^{103m}Rh based on experiments conducted at the SOLEIL synchrotron (Riffaud et al., 2016a);
- Determining the emission intensities of ^{103m}Rh derived from primary measurements (liquid scintillation) on samples of rhodium irradiated in the ISIS reactor (Riffaud et al., 2017).

Table 2
Emission intensities for ^{103m}Rh.

Data	Preliminary results	Former values	Intensity Ratio
$I_{K\alpha}$	6,89 (0.17)	6.27 (0.34)	9.9%
$I_{K\beta}$	1,36 (0.03)	1.304 (0.09)	4.3%

4.2.1. Determining the fluorescence yield of $^{93\ m}$Nb and ^{103m}Rh

The first measurements were taken on niobium and rhodium targets with a nominal thickness of 20 μm and 50 μm respectively (measured mass per unit area is 0.017042 (10) g.cm^{-2} for niobium and 0.058962 (32) g.cm^{-2} for rhodium).

The average total K fluorescence yield value obtained for niobium is ωK = 0.724 (14). For rhodium, results were obtained using an average of eight measurements with incident photons in the 26–30 keV energy range. We observed a −3.6% relative difference with LNHB nuclear database, ωK = 0.751 (4), actually used (value determinate by semi-empirical method).

The average total K fluorescence yield value obtained for rhodium is ωK = 0.814 (41). We observe a 0.6% relative difference with LNHB nuclear database ωK = 0.809 (4) actually used (value determinate by semi-empirical method).

The relative standard uncertainties of the measurements are about 2% for niobium and 5% for rhodium. The difference between these is explained by larger uncertainties on the intensity measurements of the photon flux for rhodium. The rhodium target is, in fact, thicker and significantly reduces the initial beam intensity. A new experiment has been carried out with a 6 μm sample. The analysis of these last results is under processing.

4.2.2. Determining the emission intensities of ^{103m}Rh

This experiment consisted in irradiating two rhodium samples: one pellet of pure rhodium and one sample of easily soluble rhodium chloride ($RhCl_3$). The samples were irradiated in the ISIS reactor (Foulon and X Lescop, 2011) near the LNHB building to optimise the sample recovery time

The aim of this experiment was to determine the sample activity (A) by a primary measurement through liquid scintillation. X-ray spectrometry was then conducted on the same sample, allowing the emission intensity (I) to be derived from:

$$I = \frac{A}{\tau . \varepsilon} \tag{4}$$

εFull-energy peak efficiency at energy "E_i"
τCounting rate under the full-energy peak "E_i" (s^{-1})

Preliminary experimental results are given in Table 2 and compared with tabulated data.

Preliminary results show respectively 10% and 4.5% relative deviation for Kα and Kβ rays. Associated uncertainties are strongly

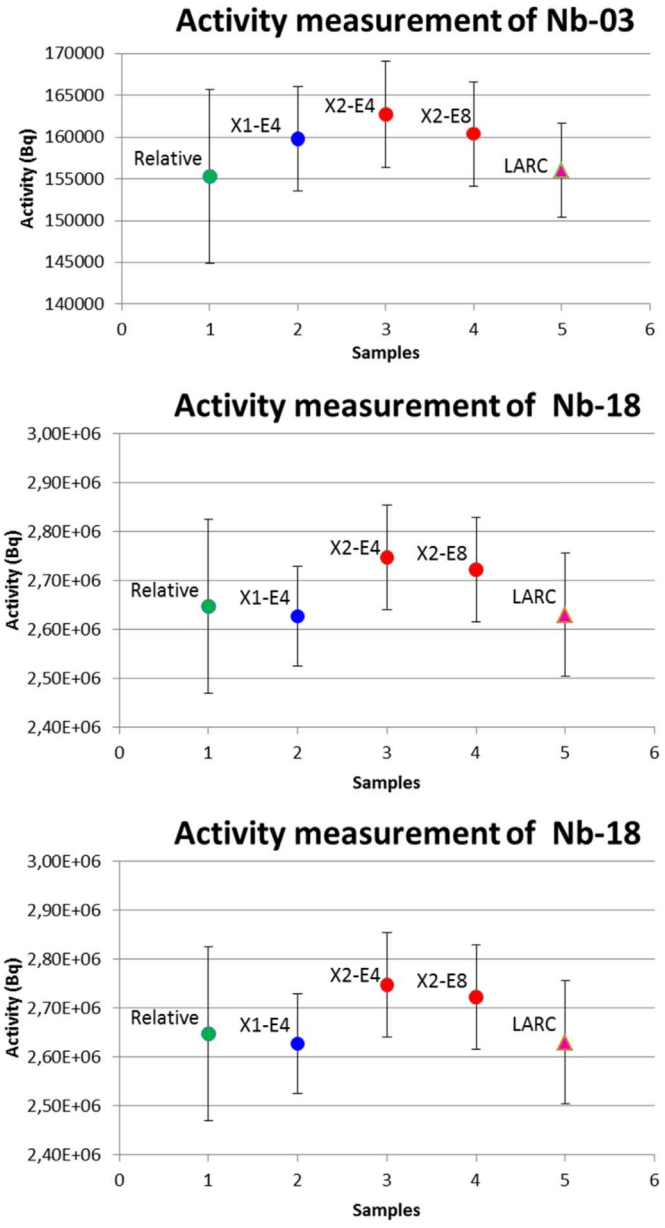

Fig. 5. Comparison of niobium activity measurements ($k=1$) using 3 different techniques.

reduced (2% instead of 5–7%). This evolution is mainly due to the improvement of absolute activity measurement techniques, especially the ratio of triple coincidences. Final results are presented in (Riffaud et al., 2017).

4.3. Experimental validation of self-attenuation corrections in ^{93m}Nb

This experimental validation consists in measuring the $^{93\,m}$Nb activity of a solid niobium sample, then dissolving this sample and measuring the activity of the liquid sample.

Given that the liquid sample is not affected by self-attenuation corrections, the comparison of these results allows the self-attenuation corrections applied to the solid sample measurements to be validated.

Three dosimeters irradiated in the French irradiation reactor OSIRIS (operated at 70 MWth) several years ago were chosen for this validation so that they were not affected by fluorescence effects.

The solid dosimeter measurements were taken by the MADERE platform on relative method and by efficiency calibration curve with the 3 existing counting geometries (X1-E4, ×2-E4 and ×2-E8). The self-attenuation correction applied to these geometries was derived by analytical calculation (IV.1.2).

The samples were dissolved by the LARC laboratory in a cold HF/HNO_3 mixture. The dissolution efficiency was determined by measurements of niobium concentration in solution by ICP/MS (Song et al., 2006). A 100% recovery efficiency was obtained for each dosimeter within the measurement uncertainties.

After dissolution, the samples are measured by X-ray spectrometry.

All the activity measurements determined by Eq. (1) and Eq. (2) are shown in Fig. 5.

The results allowed the following observations:

- The measurement based on an efficiency calibration curve technique is substantially more precise (3.0%) than the relative measurement method (6.6%), thus illustrating the benefit of extending the measurement technique based on an efficiency calibration curve - originally developed for rhodium - to niobium. It also involves better uncertainty levels than measurements taken in solution (5.0%).
- Measurement results are consistent with each other.
- The relatively high uncertainties on the radionuclides decay data have a subsequent impact on activity measurements and make it more difficult to detect any subtle inconsistencies between methods.

These results represent an initial experimental validation of self-attenuation corrections applied to niobium samples. The current re-evaluation of nuclear data on emission intensities will considerably reduce the uncertainty of results obtained and will therefore necessitate re-validation of these results.

4.4. Experimental validation of self-attenuation corrections in ^{103m}Rh

This experimental validation aims to compare measurements on rhodium samples of different thicknesses activated in an equivalent neutron flux. These experiments were carried out in the EOLE (Thiollay et al., 2014) mock-up reactor (CEA/Cadarache). An initial experiment consisted in irradiating two sets of three dosimeters, 50 μm, 12 μm and

Table 3
Comparison of rhodium dosimeter activity irradiated in two symmetrical locations.

Dosimeter	Thickness (μm)	Self-abs. Kα and Kβ	Mass (mg)	Activity (Bq/mg)	Uncertainty ($k=1$)	Difference with the reference activity (%)
Rh-63	50.93	1.584 1.398	31.769	1.151E04	4.9%	Reference
Rh-12-03	11.48	1.117 1.082	7.164	1.099E04	4.5%	−4.5%
Rh-6-03	5.64	1.057 1.040	3.518	1.094E04	4.5%	−5.0%
Rh-59	51.22	1.588 1.401	31.951	1.122E04	4.9%	Reference
Rh-12-02	11.35	1.116 1.081	7.081	1.112E04	4.5%	−0.9%
Rh-6-01	5.82	1.058 1.041	3.631	1.087E04	4.5%	−3.1%

Table 4
Comparison of the activity of rhodium dosimeters of different thicknesses.

Dosimeter	Thickness (μm)	Self-abs. K_α and K_β	Mass (mg)	Activity (Bq/mg)	Uncertainty ($k=1$)	Activity Ratio (%)
Rh – 59-X1	51.20	1.588 1.400	31.941	6.206E3	4.9%	Reference
Rh – 59-X2	51.20	1.588 1.400	31.941	6.212E3	4.9%	−0.1%
Rh – 60-X1	50.23	1.575 1.392	31.331	6.224E3	4.9%	−0.3%
Rh-12–04-X1	11.66	1.119 1.084	7.274	6.053E3	4.5%	2.2%
Rh-06–04-X2	5.74	1.058 1.041	3.581	6.068E3	4.5%	2.5%

6 μm thick, respectively, in two symmetrical locations, with comparable neutron flux conditions. The neutron flux was identical in both locations. Measurements were taken on two distinct measuring channels with similar geometry (about 3 cm). The results obtained (Eq. (2)) are shown in Table 3 indicate an agreement better than 5% lower than the measurement uncertainties.

The second experiment compared the mass activity of rhodium samples (50 μm, 12 μm and 6 μm nominal thicknesses) which were irradiated simultaneously at the same location, and thus at an identical neutron flux.

Measurements were taken on two distinct measuring channels with similar geometry (around 3 cm). The results obtained (Table 4) indicate an agreement of about 2.5%, lower than the measurement uncertainties. The self-attenuation coefficients used for the K_α and K_β lines are consolidated. However, the high uncertainties on $^{103\,m}$Rh emission intensities prevent the detection of any potential effects of low amplitude.

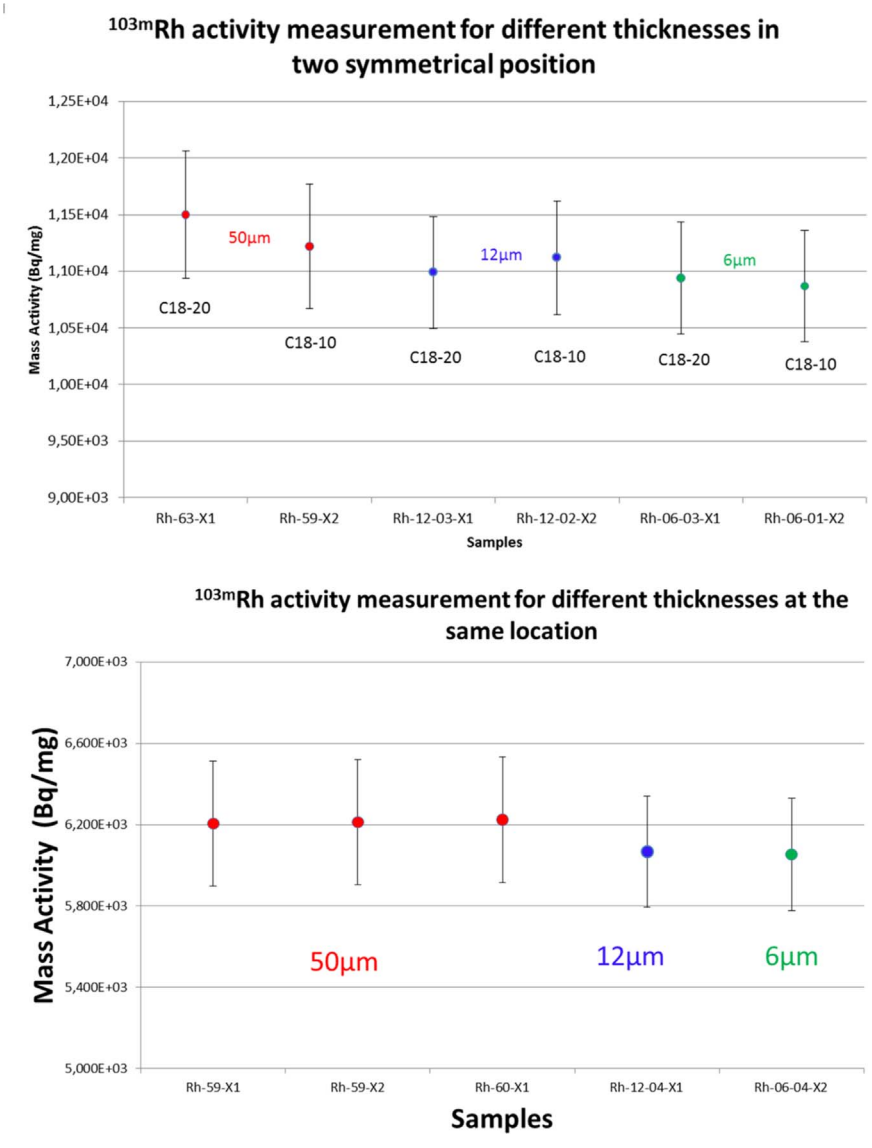

Fig. 6. $^{103\,m}$Rh specific activity ($k=1$) for dosimeters irradiated at the same location.

For each of these situations, the mass activities obtained are consistent regardless of dosimeter thickness: the differences of 1–5% are within the experimental uncertainty at $k = 1$. This experiment in EOLE has therefore enabled the experimental validation of the self-attenuation corrections used on the MADERE platform, which represent the largest correction factor equal to more than 50% for the most frequently used 50 μm dosimeters.

At this stage, use of the analytical formula to determine self-attenuation corrections is sufficient in the MADERE platform measurement conditions.

Nevertheless, Table 4 show a systematic increase in activity for the thickest dosimeters (see Fig. 6). These effects are slight and remain below the measurement uncertainties derived from the emission probability uncertainties of ^{103m}Rh.

However, a more precise re-evaluation of these emission probabilities could highlight a slight under estimation of the self-attenuation coefficient for rhodium.

5. Conclusions

Measuring the activity of solid samples by X-ray spectrometry is a complex matter. It involves several key parameters, some of which are outdated, others about which little is known. This study is essential for improving the precision of these measurements, particularly as they apply to rhodium and niobium dosimeters, both of which are fast neutron flux indicators. These measurements will enable the computational codes used in nuclear reactors to be validated in order to characterize not only the neutron flux around 1 MeV more precisely but also the resulting damage on materials used in the nuclear industry.

The aim is to calculate self-attenuation, fluorescence correction factors and emission intensities for ^{93m}Nb and ^{103m}Rh in order to obtain nuclear data with an uncertainty of less than 2%. Existing modelling tools allow the calculation of these parameters to be refined provided that the geometric data are absolutely controlled, particularly the precise characteristics of the detector. An upcoming experiment will enable comparative measurements to be taken of niobium samples, irradiated in the same flux with and without a thermal neutron filter, in order to eliminate fluorescence corrections.

Self-attenuation correction factors were experimentally validated throughout activity measurements for niobium and rhodium. Preliminary results for ^{103m}Rh and ^{93m}Nb emission intensities are given with 2% uncertainties, corresponding to the targeted goal. But a Monte-Carlo simulation is necessary in order to fully take advantage of this nuclear data improvement.

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