IMPROVEMENT OF ^{93m}Nb AND ^{103m}Rh MEASUREMENT METHODOLOGY FOR REACTOR DOSIMETRY

Link to LNHB posters

J. Riffaud[1], M.-C. Lépy[1], C. Domergue[2], J.M. Girard[2],C. Destouches[2], H. Philibert[2]

[1]CEA, LIST, Laboratoire National Henri Becquerel (LNE-LNHB), CEA-Saclay 91191 Gif-sur-Yvette Cedex, France

[2]CEA, DEN, DER, Laboratoire de Dosimétrie, Capteurs et Instrumentation (LDCI), Cadarache, 13108 Saint Paul-lez-Durance, France

Context

- Reactor dosimeters are small metal samples irradiated and activated by neutrons
- They allow: characterization of the neutron flux and energy distribution
  validation of neutron codes
  assessment of reactor vessel aging
- Each of these dosimeters has a specific neutron energy region (see table 1)
- Nb and Rh: information on neutrons with energies around 1 MeV
- Reactions of interest in this work: ^{93}Nb(n,n')^{93m}Nb and ^{103}Rh(n,n')^{103m}Rh
- Activity measurement conventionally performed by X-ray spectrometry with HPGe detector
- Currently, measurements have significant deviations (> 5%) with neutron codes and large uncertainties (> 6%)

Table 1: Dosimeter characteristics

Dosimeter	Energy of emitted photons (keV)	Neutron energy region
^{54}Fe(n,p)^{54}Mn	834.85 (γ)	Fast (3.1 MeV)
^{58}Ni(n,p)^{58}Co	810.76 (γ)	Fast (2.8 MeV)
^{197}Au(n, γ)^{198}Au	441.8 (γ)	Thermal
^{93}Nb(n,n')^{93m}Nb	16.59 (XK_α) and 18.67 (XK_β)	Fast (1 MeV)
^{103}Rh(n,n')^{103m}Rh	20.17 (XK_α) and 22.84 (XK_β)	Fast (1 MeV)

Dosimeter activity measurement

- ^{93m}Nb and ^{103m}Rh de-excite to their ground states via isomeric transitions
- Transition highly converted → emission of X-ray photons
- Spectrometer must be calibrated in low-energy range (< 100 keV)

Detector calibration

- Experimental detection efficiency established for a reference geometry
- Use of different standard point sources:
  ^{88}Y, ^{109}Cd, ^{129}I, ^{133}Ba, ^{137}Cs, ^{152}Eu, ^{207}Bi, ^{210}Pb, ^{241}Am
- Fit with least squares method → Efficiency value for any energy
- Use of a poly-logarithmic function (see figure 1)

Efficiency calculation with Monte Carlo method

- Simulation of single gamma-emitting point sources for the same geometry
- Optimization of physical parameters of the germanium crystal
- Adjustment of the simulated efficiency curve with the experimental fit (see figure 2)

- Activity measured at the same distance as reference point sources
- Dosimeters cannot be considered as point sources (see figure 3):
  Nb: strip 5 mm long, 1 mm wide and 19.89 µm thick
  Rh: pellet 8 mm diameter and 50 µm thick
- Self-absorption correction calculated with PENELOPE: ~ 20% for Nb and 50% for Rh

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

N(E): number of counts under the full-energy peak with energy "*E*"
I(E): emission intensity of the energy line "*E*"
ε(E): detection efficiency at the energy "*E*"
t: duration of the measurement
C: correction factors (self-absorption, decay, self-fluorescence, …)

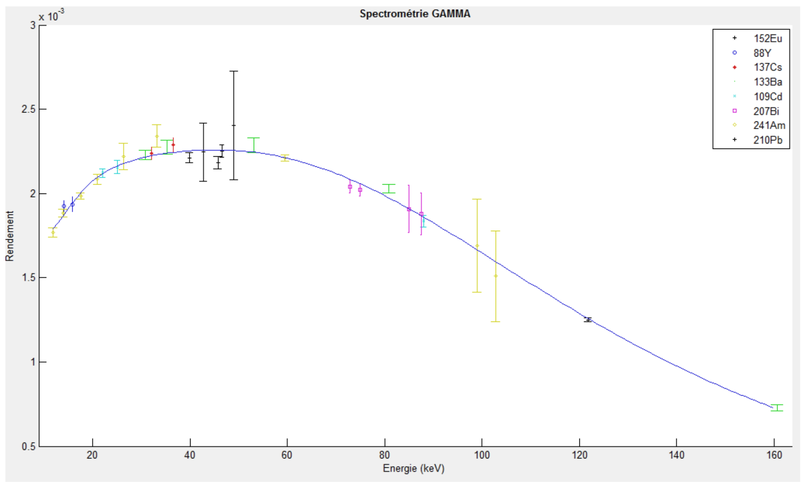

Figure 1: Experimental efficiency calibration: experimental values and fitted curve

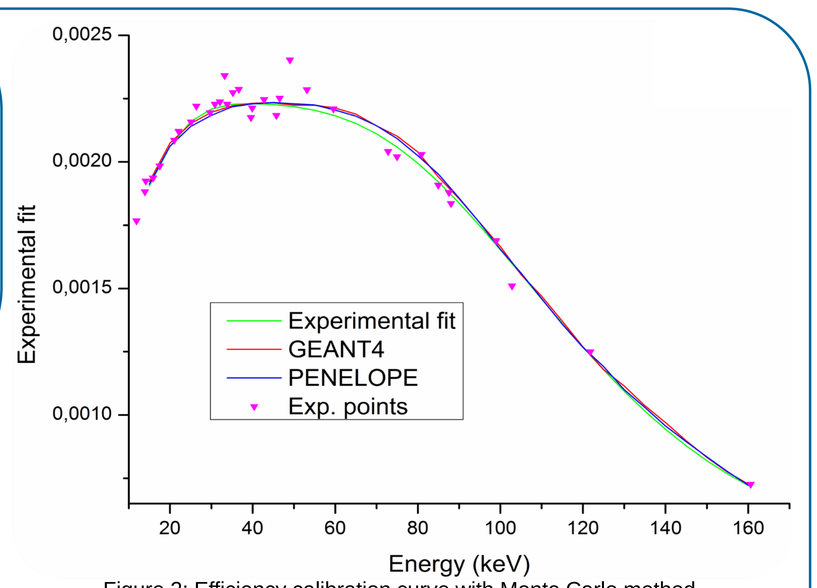

Figure 2: Efficiency calibration curve with Monte Carlo method

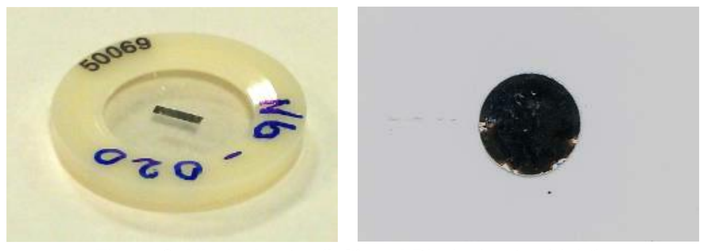

Figure 3: Nb dosimeter (left) and Rh dosimeter (right)

Self-fluorescence correction

- Niobium dosimeters are not 100% pure, other elements are present, such as Mo, W, Ta, Fe and Ni
- These impurities and other isotopes of Nb are activated during the dosimeter irradiation (see figure 4)
- ^{92}Nb, ^{94}Nb, ^{95}Nb and ^{182}Ta are still present during the activity measurement
- Photons from the decay of these impurities (blue arrows) can interact with Nb atoms by photoionization
- The consecutive electronic rearrangement produces the same K X-ray in ^{93}Nb (red arrows) as in ^{93m}Nb (green arrows)
  → Increase of the number of counts in the full-energy peaks of ^{93m}Nb
  Overestimated value of the dosimeter activity
- Corrective factors must be calculated to derive the true activity of ^{93m}Nb
- Self-fluorescence coefficients were calculated analytically in 1972 (see table 2):
  Calculated only for main energy lines
  Do not take into account:
  → The electron interactions
  → The self-absorption of Nb X-rays in the dosimeter
- Monte Carlo method applied to determine the contribution of these impurities in the dosimeter spectra

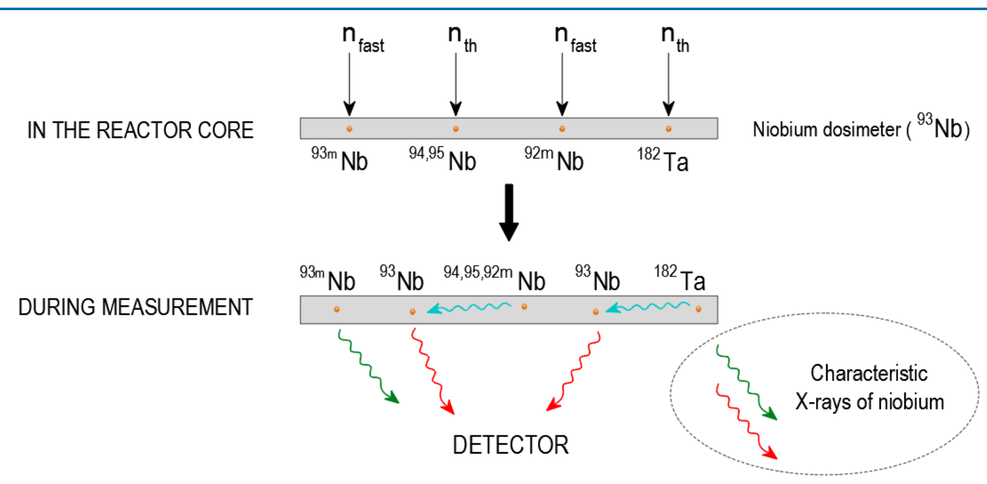

Figure 4: Self-fluorescence in niobium dosimeters

Table 2: Self-fluorescence coefficients used since 1972

Impurity	^{92m}Nb	^{94}Nb	^{95}Nb	^{182}Ta
Self-fluorescence coefficients ($s^{-1}.Bq^{-1}$)	0.021	0.0055	0.0033	0.077

Monte Carlo simulation and results

GEANT4 simulation

- Same detector geometry as for efficiency calculation and Nb volume source
- Inclusion of the impurities' radioactive decay from the decay schemes (see figures 5 and 6)
  → Use of PenNuc files (see figure 7)
  → Takes into account the branching ratios, emission intensities and internal conversion
  → Emission of photons and electrons with random position in Nb and random direction within 4π
- Simulated radionuclides: ^{92m}Nb, ^{94}Nb, ^{95}Nb, ^{182}Ta and ^{60}Co

Results

- Results obtained for the same source geometry are rather close to the coefficients found in 1972 (see table 3)

Table 3: Results of ^{95}Nb and ^{182}Ta impurities in the Nb dosimeter

Impurity	^{95}Nb	^{182}Ta
Self-fluorescence coefficients ($s^{-1}.Bq^{-1}$)	0.0030	0.065

- The ^{182}Ta impurity causes between 0.5% and 7% of the self-fluorescence in the Nb, of which 20% is due to electron interactions
- The ^{182}Ta coefficient calculated in table 2 does not take into account electron interactions (20% increase of the self-fluorescence) and X-ray self-absorption in the dosimeter (20% decrease)

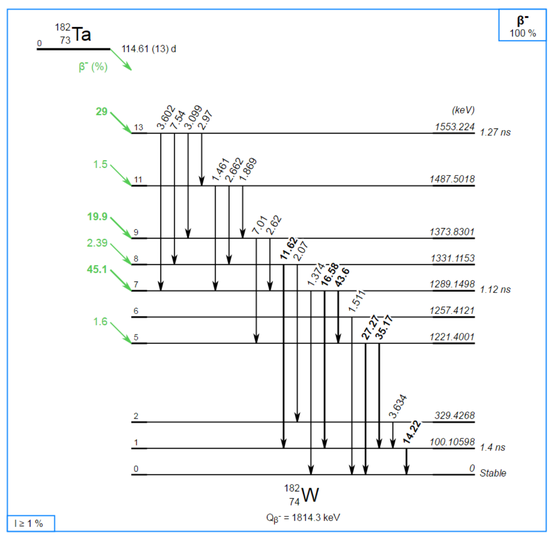

Figure 5: Simplified decay scheme of the ^{182}Ta

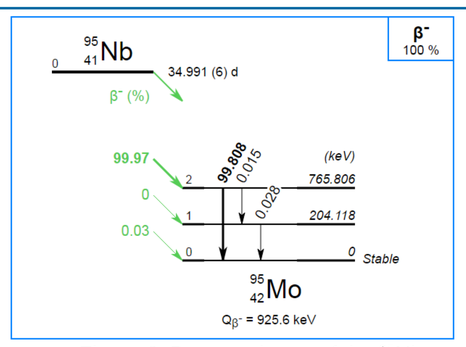

Figure 6: Decay scheme of the ^{95}Nb

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PAR NB95
AZP 95 ; 41
NDA 1
COM ***************** Evaluation Date: 27/07/1998 ****************
COM ====================== Daughter Separator ====================
DAU MO95
DDE 1 ;  ; 2 ; 2
Q   925.6 ; 0.5
COM ---------------------- Branch Separator ----------------------
BEM 0.9997 ; 0.00006 ; 2 ; 159.8 ; 0.5 ; 0
COM ---------------------- Branch Separator ----------------------
COM ---------------------- Branch Separator ----------------------
BEM 0.0003 ; 0.00005 ; 0 ; 925.6 ; 0.5 ; 1
COM ------------- Level Separator - T1/2 in seconds -------------
LED 765.806 ; 0.006 ; 6 ; 4.4E-12 ; 7.E-13 ; 2
GA  0.00015 ; 0.00003 ; 561.88 ; 0.02 ; 1
EK  0.00000045 ; 0.00000009 ; 541.88 ; 0.02 ; 1
EL  0.000000053 ; 0.000000011 ; 559.21 ; 0.02 ; 1
GA  0.99808 ; 0.00007 ; 765.803 ; 0.006 ; 0
EK  0.001288 ; 0.00004 ; 745.806 ; 0.006 ; 0
EL  0.0001447 ; 0.000004 ; 763.136 ; 0.006 ; 0
COM ------------- Level Separator - T1/2 in seconds -------------
LED 204.118 ; 0.009 ; 3 ; 7.51E-10 ; 9.E-12 ; 1
GA  0.00028 ; 0.00008 ; 204.117 ; 0.002 ; 0
EK  0.0000126 ; 0.0000037 ; 184.1175 ; 0.002 ; 0
EL  0.00000162 ; 0.00000048 ; 201.4467 ; 0.002 ; 0
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Figure 7: Example of a PenNuc file with ^{95}Nb

Next steps

- Similar approach for Rh dosimeter impurities (iridium and platinum)
- Experimental validation of the simulation and assessment of uncertainties

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