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Presentation

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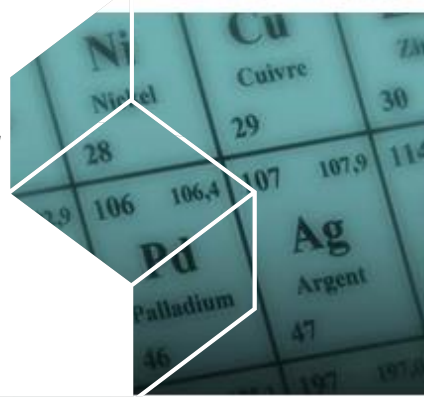
Fluorescence correction for activity measurement of ^{93m}Nb in niobium dosimeters: calculation and experimental validation

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Outline

Introduction

X-ray spectrometry

- Efficiency calibration

- Processing of X-ray spectra

- Self-attenuation correction

Fluorescence effects

- Corrective factors

Experimental validation

- Spectrometry measurements

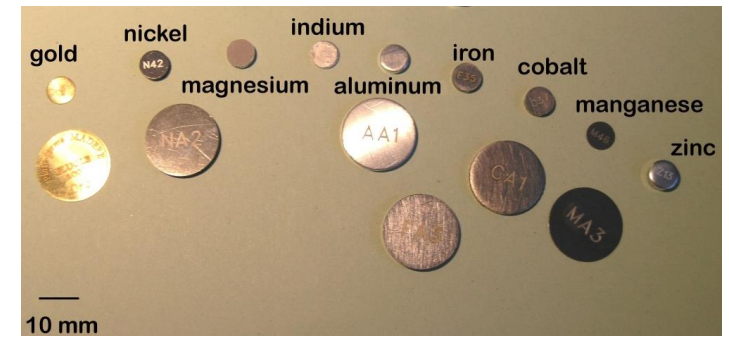
- Activity results

Conclusion

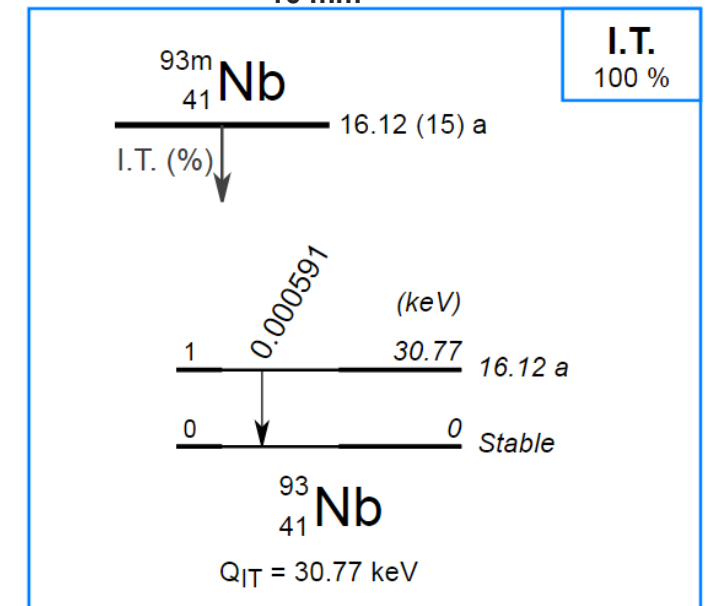
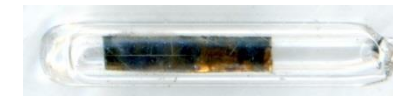
Introduction

- Niobium dosimeters are used in reactors to characterise neutron fluxes in the energy range around **1 MeV**.
- Information based on the activity of ^{93m}Nb resulting from the activation reaction $^{93}\text{Nb}(n, n')$.
- ^{93m}Nb decays by isomeric transition with a strong internal conversion coefficient.
- This decay results mainly in emission of niobium K X-rays which are used to determine the activity of ^{93m}Nb by:
 - **direct** measurement (solid sample) using **X-ray spectrometry**,
 - after sample dissolution, using X-ray counting or liquid scintillation counting.

| Half-life (years) | Type of emission | Energy (keV) | Intensity (%) |
|-------------------|------------------|--------------|---------------|
| 16.12 (15) | Gamma | 30.77 | 0.000591 (9) |
| | XK α | 16.58 | 9.66 (17) |
| | XK β | 18.67 | 1.89 (4) |



niobium



^{93m}Nb decay scheme

Introduction

- Direct measurement : **no preparation** of the sample

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

A: sample activity

N(E): Net peak area

I(E): photon emission intensity

ε(E): detection efficiency

t: counting time (live time)

C_i: corrective factors

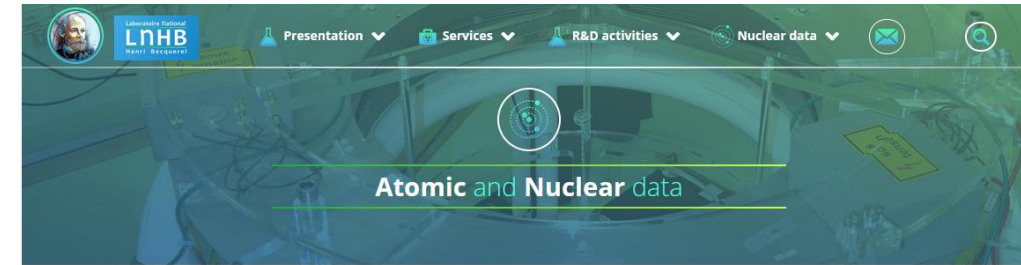
- Some difficulties for X-ray spectrometry:

- Efficiency calibration $\varepsilon(E)$ and spectra processing
- Self-attenuation within the dosimeter volume
- Fluorescence effects

Radionuclide decay data: DDEP

<http://www.lnhb.fr/nuclear-data/>

<http://www.lnhb.fr/nuclear-data/module-lara/>



Many applications, such as applied research or detector calibration, require knowledge of the atomic and nuclear data that follow radioactive decay, e.g. half-life, decay modes and branching ratios, the energies and intensities of the various emissions, etc.

In order to provide users with carefully recommended data, an international working group (Decay Data Evaluation Project, **DDEP**) was created in 1995. The update of the recommended data pages is performed by the Laboratoire National Henri Becquerel.

More specifically for users of alpha and gamma spectrometry, we have also developed **Nucléide-Lara**.

The online application makes it possible to consult and query the decay data for more than 400 radionuclides (half-life, decay mode, emission energy and intensity, decay scheme...).

Below are the **tables of recommended data** for more than 200 radionuclides with detailed comments describing how these values were obtained.

Nucléide-Lara Application

Tables of recommended data

Nucléide - Lara
Library for gamma and alpha emissions

Nuclide list:
 93Nb-M
 93Mo
 93Mo-M
 94Sr
 94Y

Nuclide, element or mass number search:

 (e.g.: 57Co, Co-57, Co, 57)

Energy threshold (keV):

Intensity threshold (%):

Coincidence threshold (%):

Show γ - γ coincidences

Sort by decreasing intensity

Show daughter emissions

Sort by nuclide if in chain

Display:
 Data Emissions Tools Scheme
 X Gamma Alpha

Language: EN EO FR

Nuclide search criteria
 And Or XOR

Decay mode: β^+ β^- IT α

Emissions: X Gamma Alpha

Energies (or range):
 E1: keV } And
 E2: keV } Or
 E3: keV } Or

Intensity: %

93mNb - Emissions and decay scheme

Data Emissions Tools Scheme

Data
 Element: Niobium (Z=41)
 Daughters: Nb-93 (I.T., 100%)
 Q^{γ} : 30.77 keV
 Half-life (T_{1/2}): 16.12 (15) a + 508.7 (47) 10⁶ s
 Decay constant (λ): 1.363 (13) 10⁻⁹ s⁻¹
 Specific activity (A_m): 8.82 (8) 10¹² Bq g⁻¹
 Reference: KRI - 2013
 Associated data files: [Table](#) - [Comments](#) - [ENSDF](#) - [FenNuc](#)
 Data and emissions file (ASCII text format): [Nb-93m.lara.txt](#)

Emissions
 Coincidence threshold: 10%
 Emissions (6 lines) sorted by decreasing intensity

| Energy (keV) | Intensity (%) | Type | Origin* | Levels Start* End* | Possible coincidence with (keV) / Possible sum of (levels) |
|--------------|---------------|------------------|---------|--------------------|--|
| 16.6152 (-) | 6.34 (15) | X _{Kα1} | Nb | | |
| 16.5213 (-) | 3.32 (8) | X _{Kα2} | Nb | | |
| 2.285 (-) | 2.88 (6) | X _L | Nb | | |
| 18.67 (-) | 1.64 (4) | X _{Kβ1} | Nb | | |
| 18.967 (-) | 0.246 (11) | X _{Kβ2} | Nb | | |
| 30.77 (2) | 0.00591 (9) | γ | Nb-93 | 1 0 | |

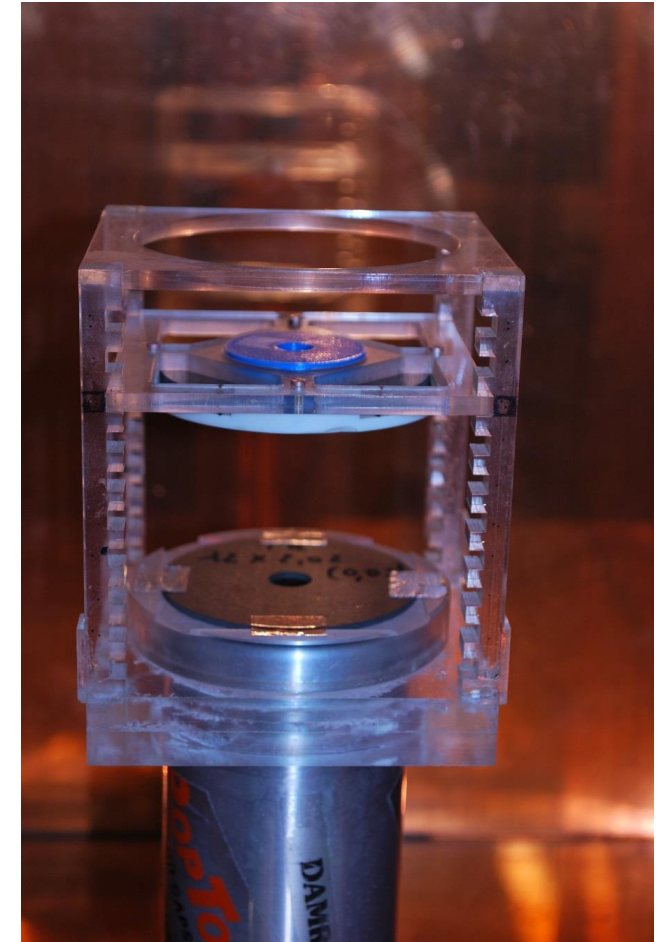
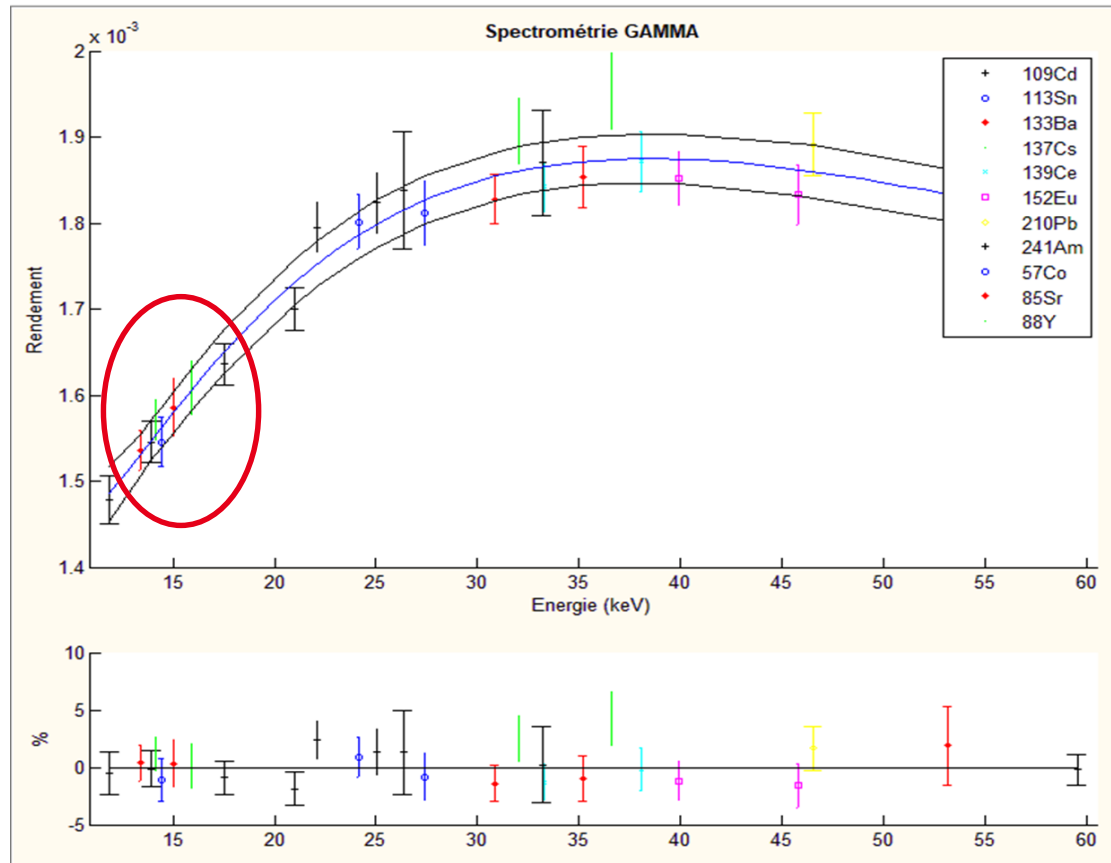
Tools
 Activity = Mass conversion: 1000 Bq = 1.133E-10 g
 Decay calculation: 1 calculation step(s)
 t₁ → t₂ = 1.612E1 → 3.224E1 a
 d₁ → d₂ = 05 | 20 | 2023 | 10:53:17 → 05 | 21 | 2023 | 10:53:17
 Nuclide (T_{1/2}) A_p A_n
 93mNb (16.12 a) 1000 500 250 Bq

X-ray spectrometry

High-purity germanium (HPGe) detector (N-type)

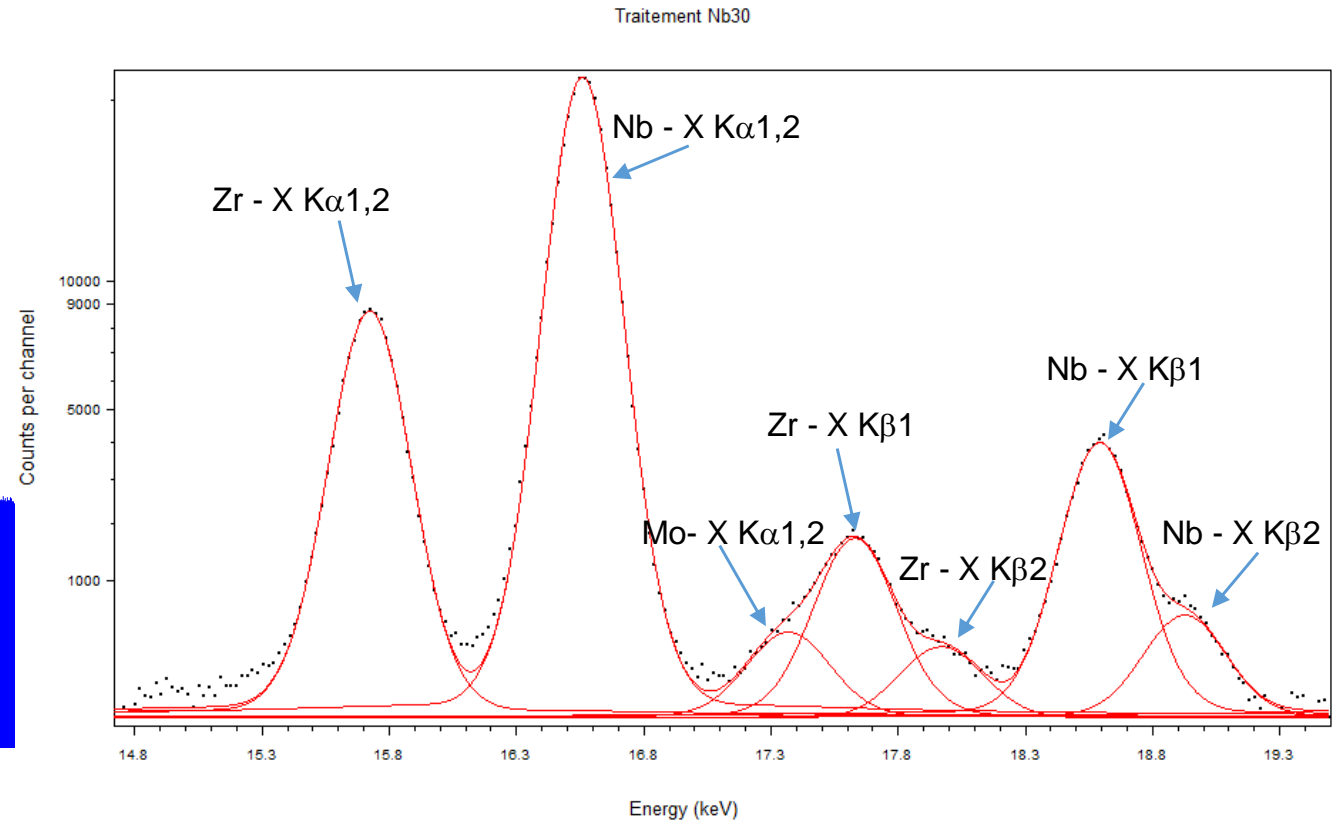
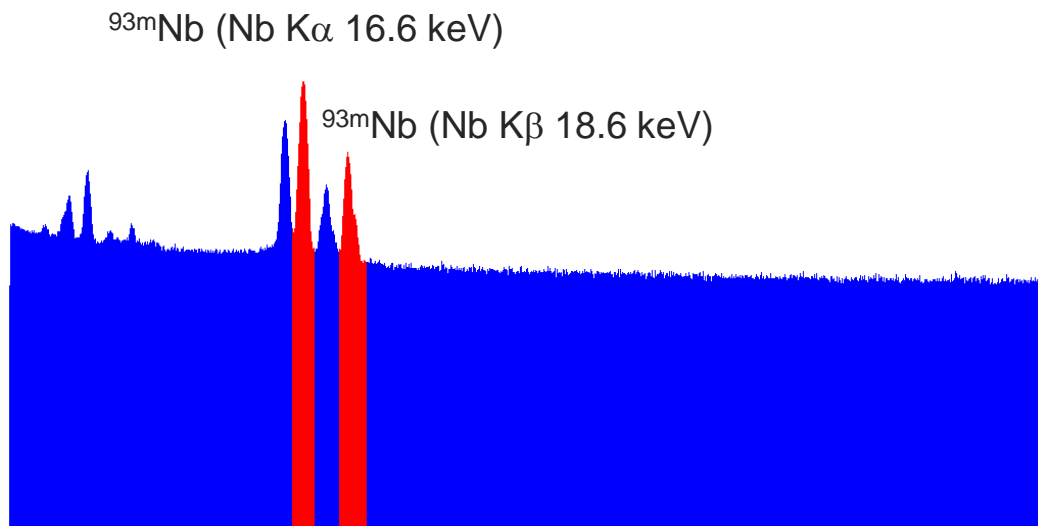
Source-to-window distance = 8 cm

Efficiency calibration using standard point sources – relative uncertainty = 1.1 %



X-ray spectrometry

Typical X-ray spectrum
Includes X-rays from Nb, Zr and Mo



COLEGRAM* software: detailed processing using Voigt function for each peak (X-ray natural linewidth)

* Mènesguen, Y., Lépy, M.-C., 2021. *COLEGRAM, a flexible user-friendly software for processing of ionizing radiation spectra*, Nuclear Inst. and Methods in Physics Research, A 1003,165341. <https://doi.org/10.1016/j.nima.2021.165341>

X-ray spectrometry

Self attenuation

The number of **existing** photons is lower than the number of **emitted** photons.

Depends on the **material** and on the **energy**

Depends on the **thickness** (x) of the dosimeter

$$\text{Approximation: } C_{att} = \frac{1 - \exp(-\mu \cdot x)}{\mu \cdot x}$$

μ : mass attenuation coefficient of Nb for the energy of emitted photons

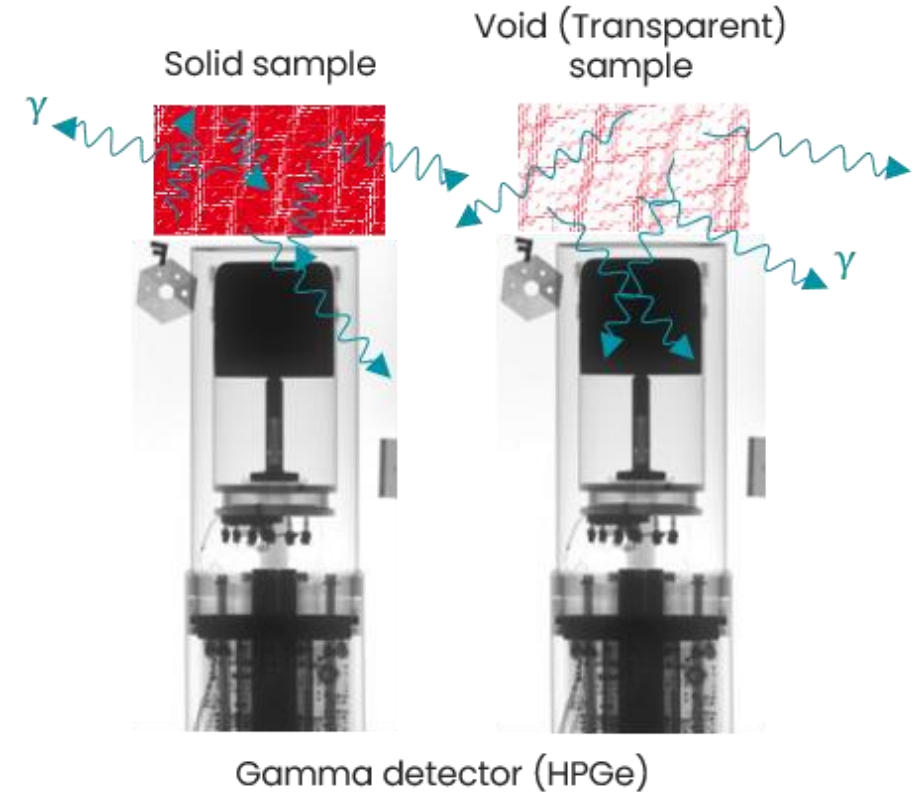
C_{att} also depends on the measurement geometry

Corrective factor ($1/C_{att}$) by dedicated software
(GESPECOR, EFFTRAN, ETNA, etc.)

Or using Monte Carlo simulation

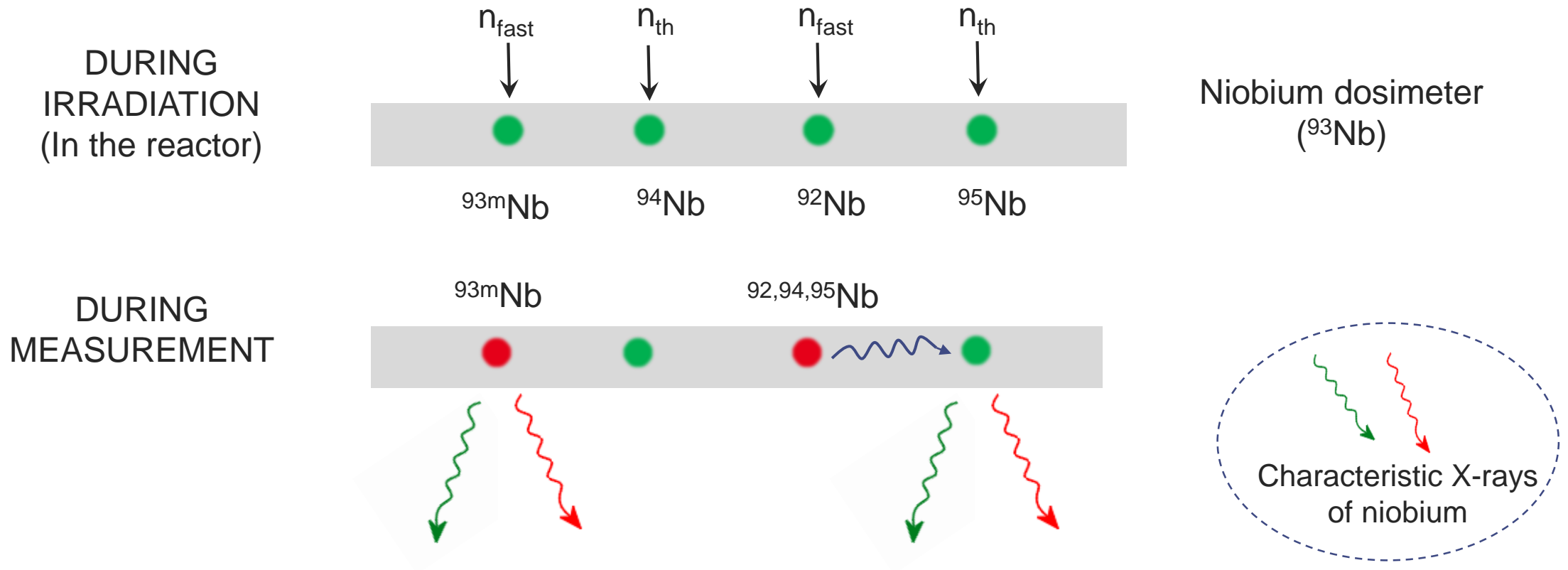
20 μm -thick Nb at 8 cm:

| Self attenuation | $K\alpha$ | $K\beta$ |
|------------------|-----------|----------|
| Strip | 0.89 | 0.93 |
| Disc | 0.90 | 0.92 |



Fluorescence corrections

1. Activation products from ^{93}Nb
(n, 2n) (n, γ), (n, p) reactions



Additional Nb X-rays due to fluorescence from activated Nb

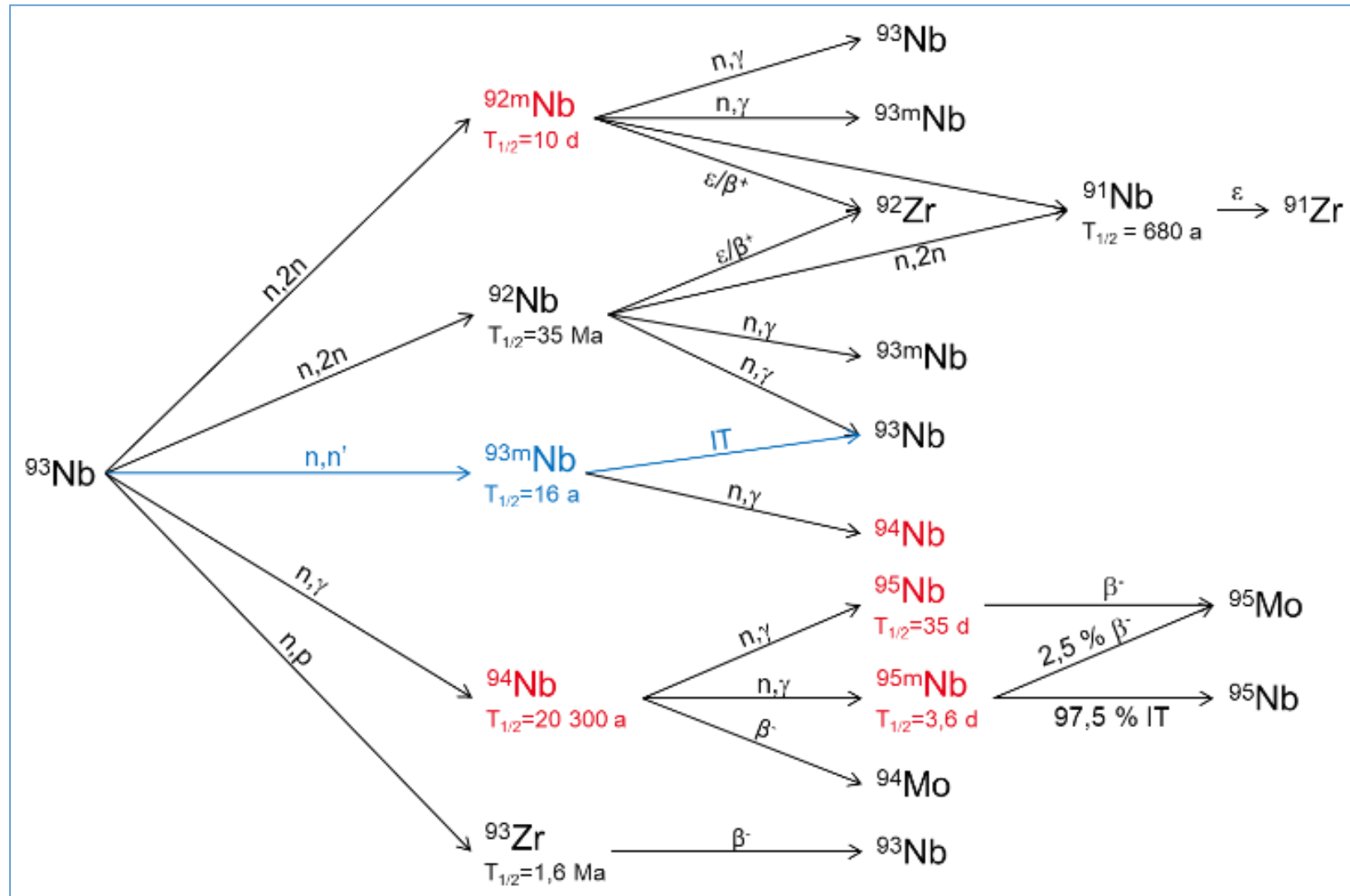
Fluorescence corrections

1. Activation products from ^{93}Nb
(n, 2n) (n, γ), (n, p) reactions

Peaks due to the decay of Nb
activation products

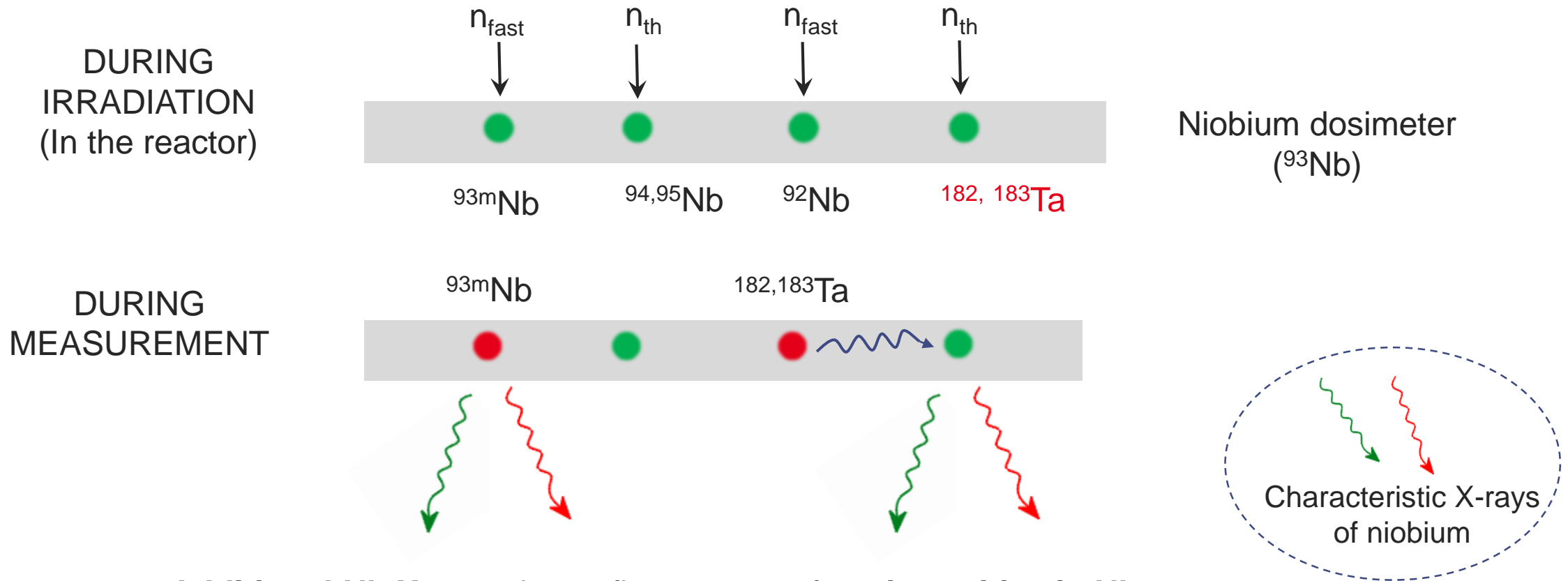
Gamma rays*
X rays (Zr, Mo)
Nb K-X rays

*Quantification using gamma-ray
spectrometry



Fluorescence corrections

2. Activation products of the impurities (Ta, Mo, W)

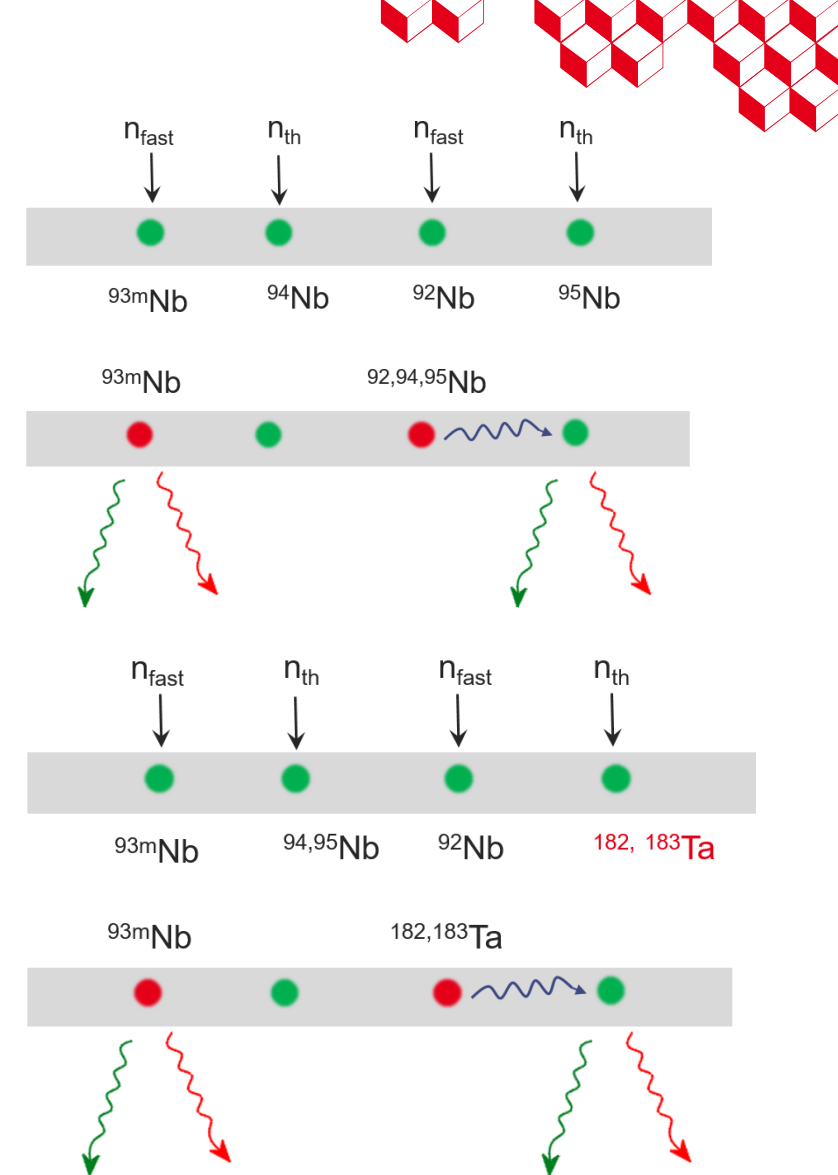


Additional Nb X-rays due to fluorescence from impurities in Nb

Fluorescence corrections

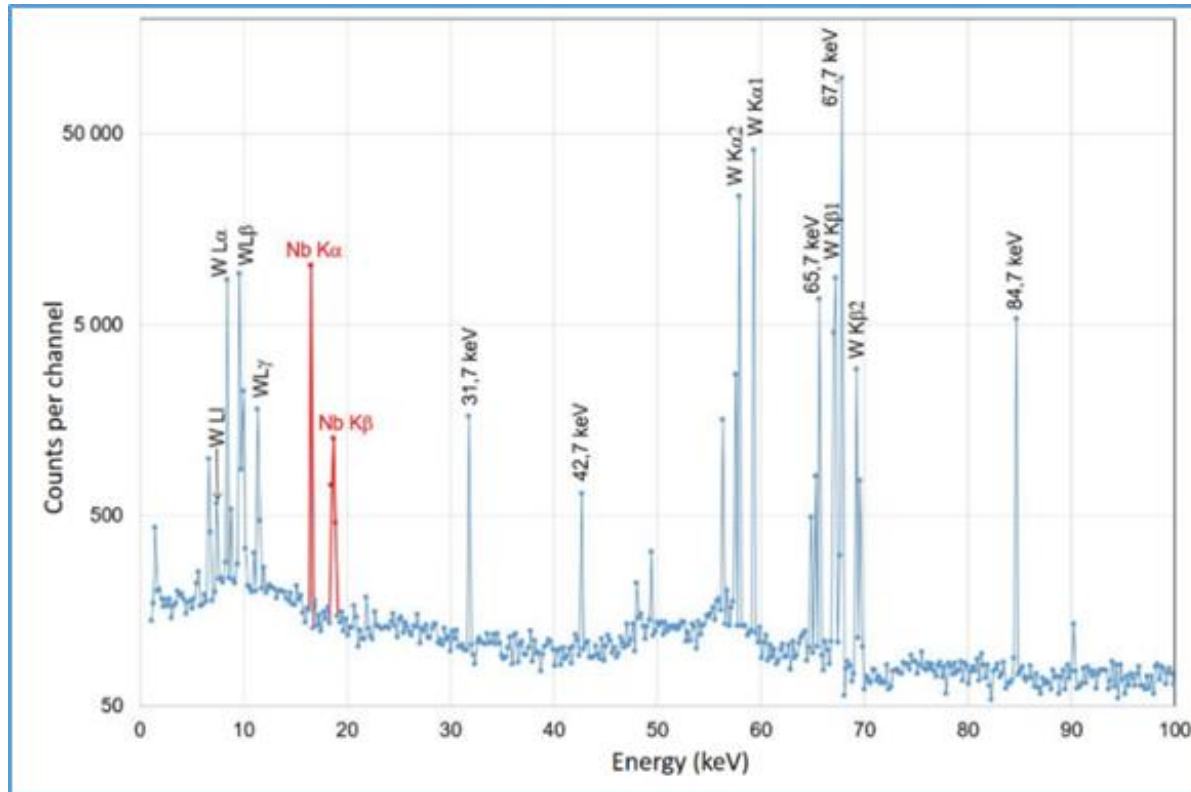
Main characteristics of ^{93m}Nb and of gamma-emitting impurities

| Radionuclide | Half-life | Energy (keV) | Emission intensity (%) | Reference |
|-------------------|--------------------|----------------------|--------------------------|-------------------------|
| ^{93m}Nb | 16.12 (15) a | 16.58 18.67 | 9.66 (17) 1.89 (4) | DDEP – KRI 2013 [4] |
| ^{92m}Nb | 10.15 (2) d | 934.44 | 99.07 (4) | NDS 66 – 1999 [3] |
| ^{94}Nb | 20.0 (24) 10^3 a | 702.62 871.09 | 97.9 (20) 99.9 (1) | NDS 44 & 66 - 1999[3] |
| ^{95}Nb | 34.991 (6) d | 765.80 | 99.808 (7) | DDEP – INEL 2002[4] |
| ^{95m}Nb | 3.61 (3) d | 235.69 | 25.1 (3) | DDEP – LPRI-INEL 202[4] |
| ^{182}Ta | 114.61 (13) d | 1 121.29 1 221.39 | 35.17 (33) 27.27 (27) | DDEP – LNHB 2011[4] |
| ^{183}Ta | 5.1 (1) d | 246.06 | 27.2 | NDS [3] |



Fluorescence corrections

Monte Carlo simulation (GEANT4 – PENELOPE)



Simulated spectrum following the decay of ^{182}Ta in a niobium dosimeter

Correction factors for fluorescence induced by gamma-emitting impurities

$$C_N(E) = \frac{1}{\varepsilon(E)} \cdot \frac{N_P}{N_T}$$

N_p : net peak area ($K\alpha$ or $K\beta$)

N_T : total number of simulated

$\varepsilon(E)$: detection efficiency

| Radio-nuclide | Corrective factor for $K\alpha$ ($\text{s}^{-1}\cdot\text{Bq}^{-1}$) | Corrective factor for $K\beta$ ($\text{s}^{-1}\cdot\text{Bq}^{-1}$) |
|--------------------------|--|---|
| $^{92\text{m}}\text{Nb}$ | $1.49 \cdot 10^{-4}$ | $2.20 \cdot 10^{-5}$ |
| ^{94}Nb | $1.00 \cdot 10^{-2}$ | $1.85 \cdot 10^{-3}$ |
| ^{95}Nb | $2.55 \cdot 10^{-3}$ | $5.5 \cdot 10^{-4}$ |
| $^{95\text{m}}\text{Nb}$ | 0.364 | 0.071 |
| ^{182}Ta | $5.42 \cdot 10^{-2}$ | $1.05 \cdot 10^{-2}$ |
| ^{183}Ta | $8.21 \cdot 10^{-2}$ | $1.48 \cdot 10^{-2}$ |

Experimental validation

Part of the European Working Group on Reactor Dosimetry Round Robin on ^{93m}Nb *

3 levels of Ta impurities (0.3 ppm, 4 ppm and 19.6 ppm) – other impurities: **Mo, W, Zr**, Fe, Ni, Cr, Co

^{93m}Nb activity of 9 dosimeters (3 strips, 6 discs) followed during 1 year

Typical dimensions :

- Strips : thickness 20 μm , length 7 mm, width 1 mm, mass 1 mg,
- Discs : thickness 20 μm , length 8 mm, mass 8.6 mg,

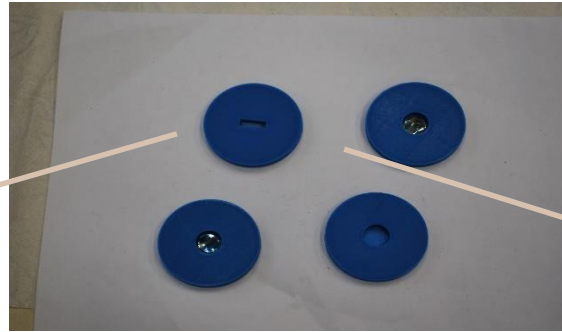
Gamma-ray spectrometry : quantification of impurities

X-ray spectrometry : ^{93m}Nb activity

* D. Thorton et al., The Second EWGRD Round Robin: Inter-Comparison of ^{93m}Nb Measurements, EPJ Web of Conferences 278, 05002 (2023)
<https://doi.org/10.1051/epjconf/202327805002>

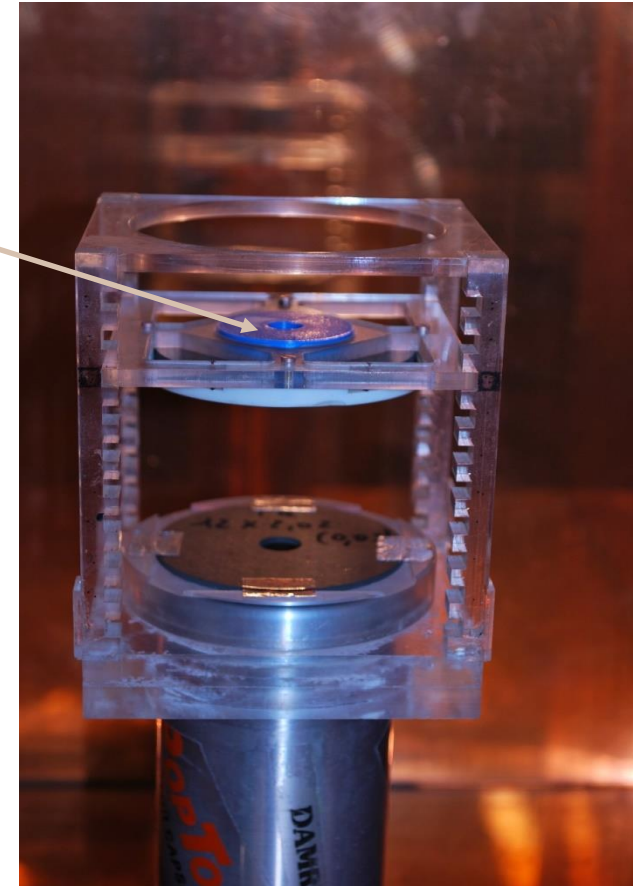
Experimental validation

^{93m}Nb activity of 9 dosimeters (3 strips, 6 discs) followed during 1 year
Gamma-ray spectrometry : quantification of impurities
X-ray spectrometry : ^{93m}Nb activity



Gamma : GeHP – sample
at 10 or 20 cm
Dead time < 5 %

X: GeHP – sample at 8 cm
Counting time : 9000 s to 63 000 s
Dead time < 1 % (strips)
< 5% (discs)



Experimental validation

Typical gamma-ray spectrum with 0.3 ppm Ta

Main peaks from:

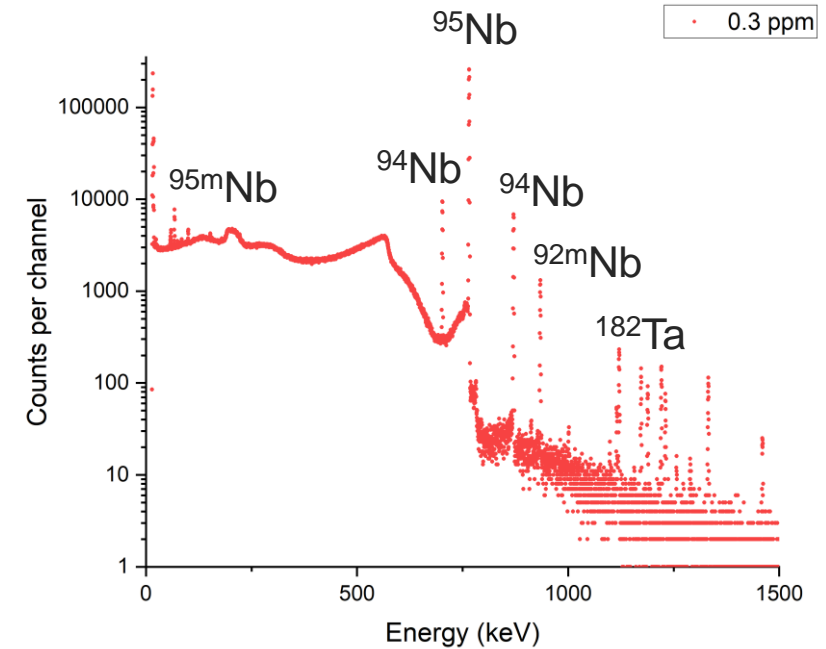
^{92m}Nb : 933 keV

^{94}Nb : 703 keV and 870 keV

^{95}Nb : 766 keV

^{95m}Nb : 236 keV

^{182}Ta : 1121 keV and 1221 keV

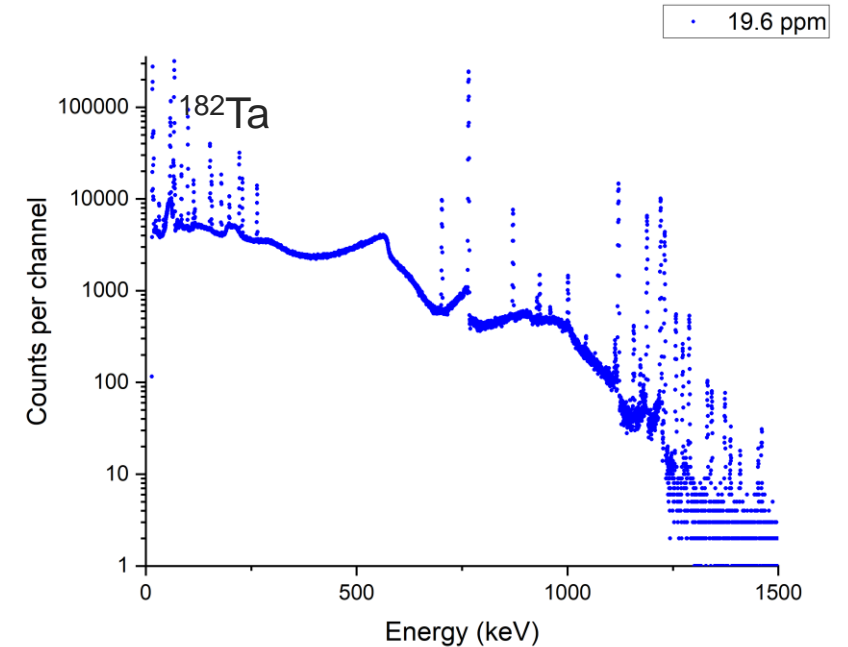


Typical gamma-ray spectrum with 19.6 ppm Ta

Additional peaks from

^{182}Ta (152 keV – 156 keV

179 keV - 229 keV - 264 keV)



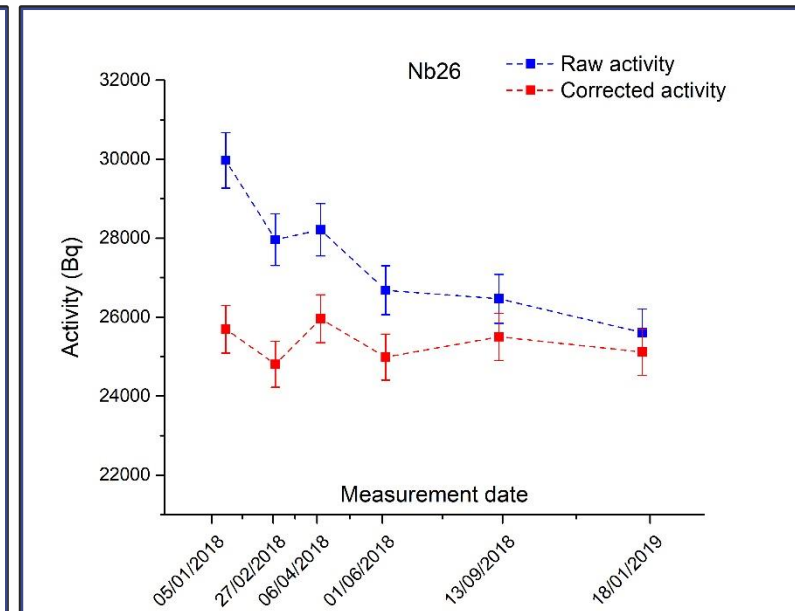
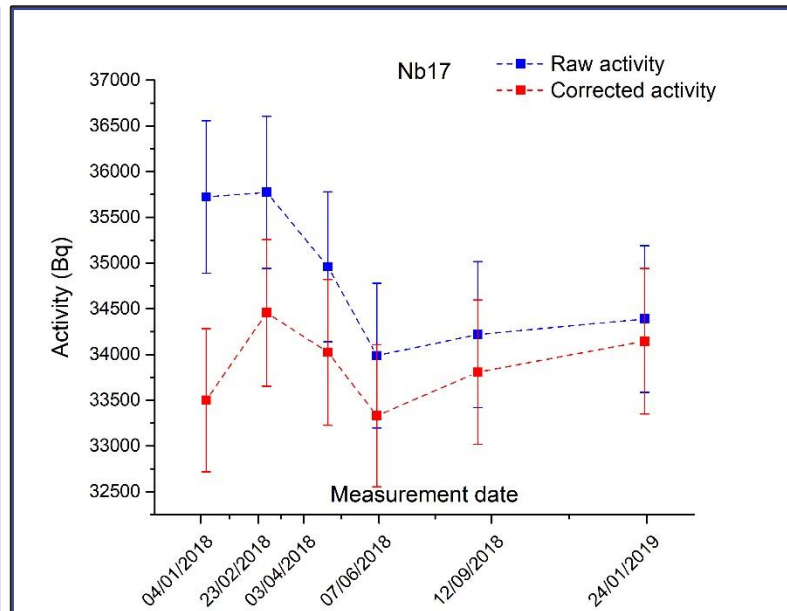
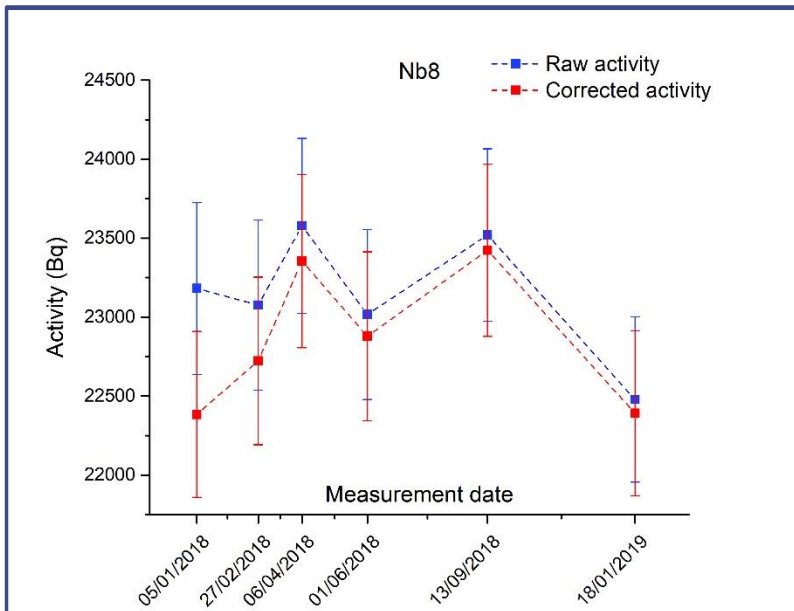
For each dosimeter, the net areas of the X-ray peaks of Nb (X-ray spectrometry) are corrected for the fluorescence induced by the quantified impurities

Experimental validation

Strips 0.3 ppm

4 ppm

19.6 ppm

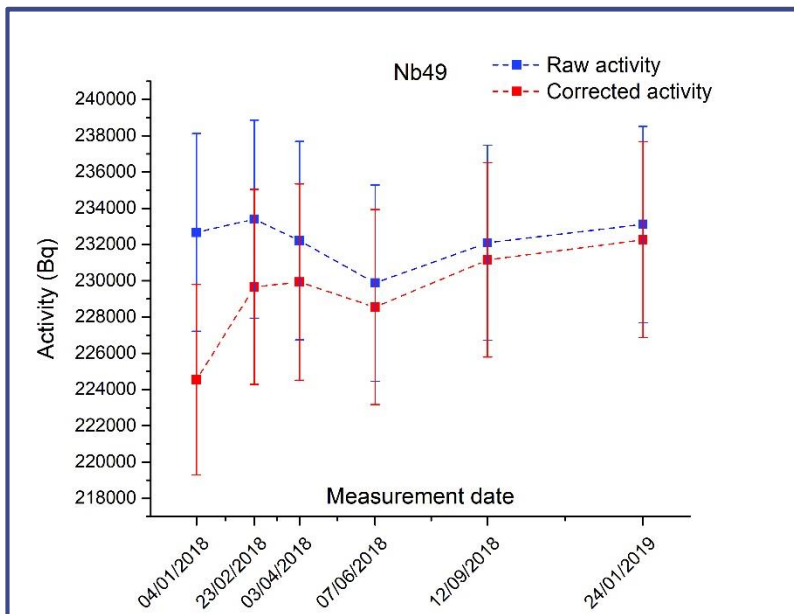


| Dosimeter number | Proportion Ta (ppm) | Raw results | | With correction | |
|------------------|---------------------|--------------------|---------------------------------|--------------------|---------------------------------|
| | | Mean activity (Bq) | Relative standard deviation (%) | Mean activity (Bq) | Relative standard deviation (%) |
| Nb8 | 0.3 | 23 280 | 1.11 | 22 860 | 1.98 |
| Nb17 | 4 | 34 840 | 2.22 | 33 880 | 1.23 |
| Nb26 | 19.6 | 27 480 | 5.67 | 25 390 | 1.90 |

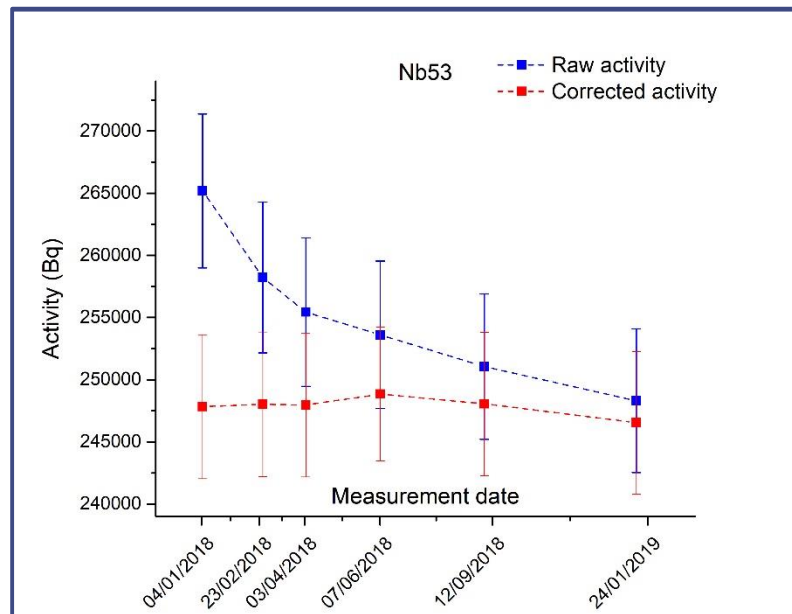
2% to 8% relative difference between the two series

Experimental validation

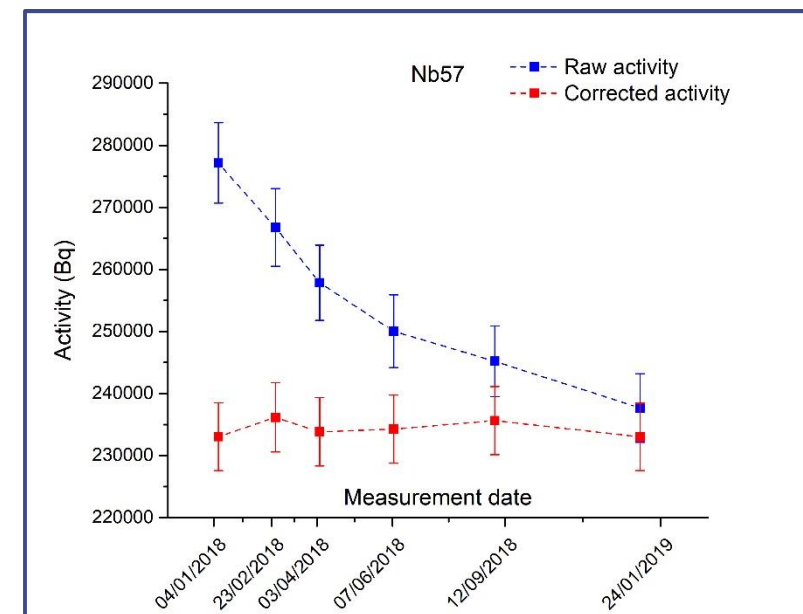
Dics 0.3 ppm



4 ppm



19.6 ppm



| Dosimeter number | Proportion Ta (ppm) | Raw results | | With correction | |
|------------------|---------------------|--------------------|---------------------------------|--------------------|---------------------------------|
| | | Mean activity (Bq) | Relative standard deviation (%) | Mean activity (Bq) | Relative standard deviation (%) |
| Nb49 | 0.3 | 232 230 | 0.54 | 229 350 | 1.17 |
| Nb53 | 4 | 255 300 | 2.32 | 247 880 | 0.30 |
| Nb57 | 19.6 | 255 770 | 5.68 | 234 330 | 0.56 |

1.2% to 8% relative difference between the two series

Conclusion

- Experimental approach to **validate fluorescence corrective factors** for activity measurement of Nb dosimeters.
- 2-step procedure:
 - Gamma-ray to quantify impurities,
 - X-ray spectrometry with corrective factors.
- Typical relative uncertainty on the activity about 2 %.
- **Direct** measurement of the activity Nb dosimeters using X-ray spectrometry.
- Possibility of obtaining **results shortly after the end of the irradiation.**

cea

Thank you for your attention

