



list
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CALIBRATION OF GAMMA-RAY SPECTROMETERS FOR GAS CONTAINERS

DE LA RECHERCHE À L'INDUSTRIE



ICRM-LLRMT – May 2-6, 2022

Marie-Christine Lépy, Sylvie Pierre, Matias Rodrigues, Benoît Sabot



Introduction

Measurement of reference activity (Kr-85, Xe-133, Xe-127, Rn-222)

Length compensated internal gas proportional counters

Alpha spectrometry of frozen source

Application to gamma-ray spectrometry

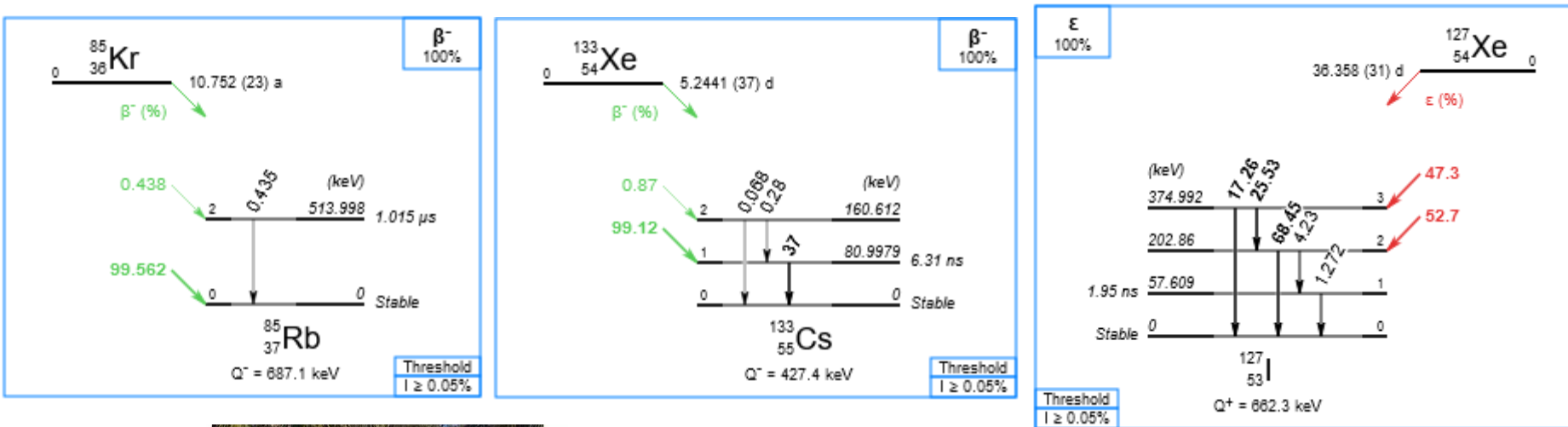
Calibration factors

Difficulties

Conclusion

Needs to quickly measure the activity of radioactive noble gases \rightarrow gamma-ray spectrometry

Example of decay schemes:



CTBTO



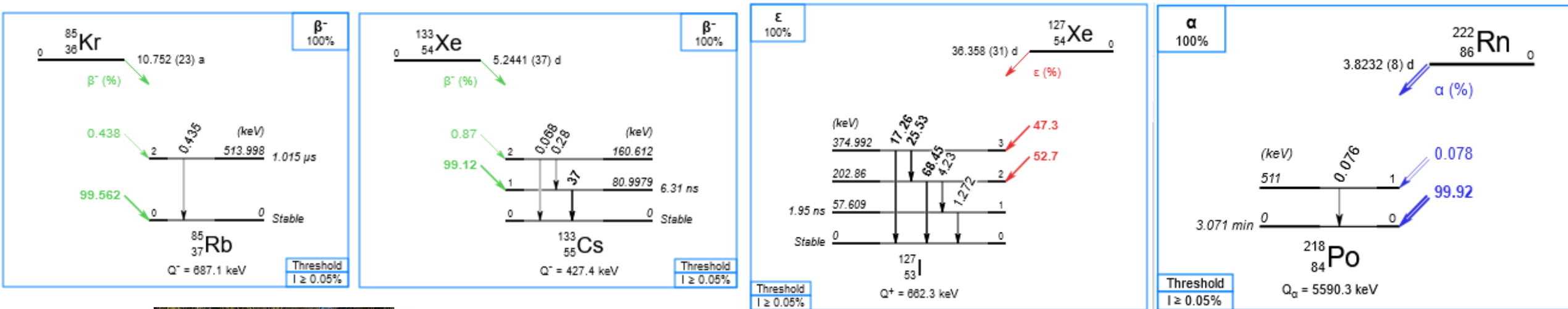
Fission products



Needs to quickly measure the activity of radioactive noble gases → gamma-ray spectrometry

Example of decay schemes:

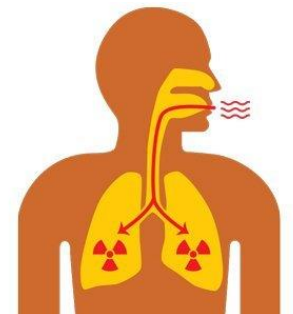
^{238}U decay chain



CTBTO

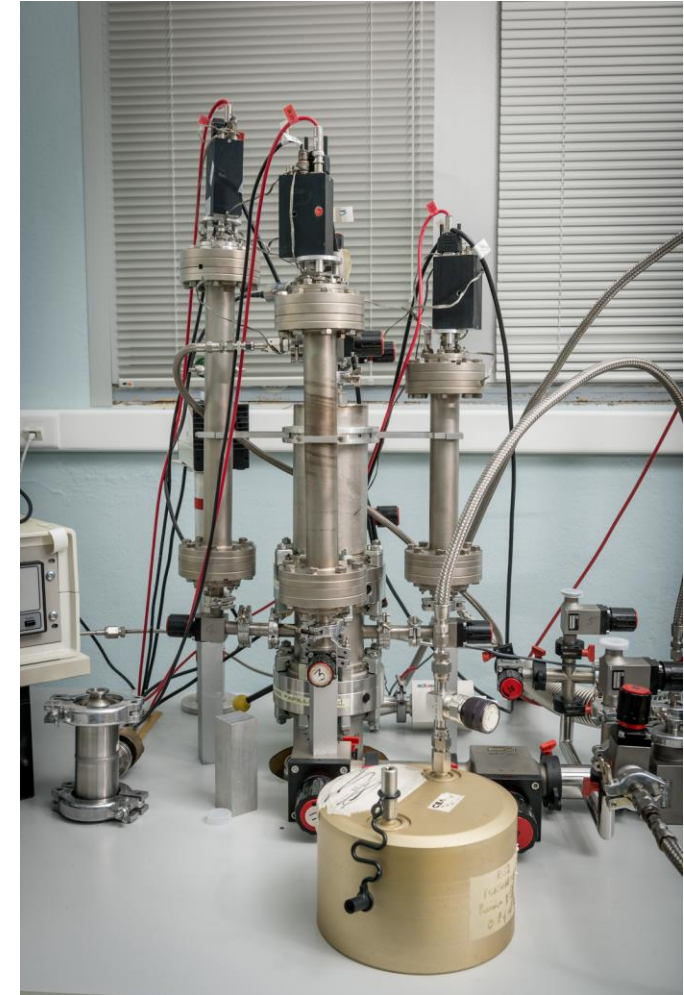


Fission products



Primary standard technique using a **length compensated triple proportional counter**, established since 1993 at LNE-LNHB:

- ✓ Mix of propane and radioactive gas;
- ✓ Require precise pressure, temperature and volume measurements;
- ✓ Three counters with same geometry, except their length;
- ✓ Differential measurement between each counter to remove the edge effects (where the electric field is not well defined).



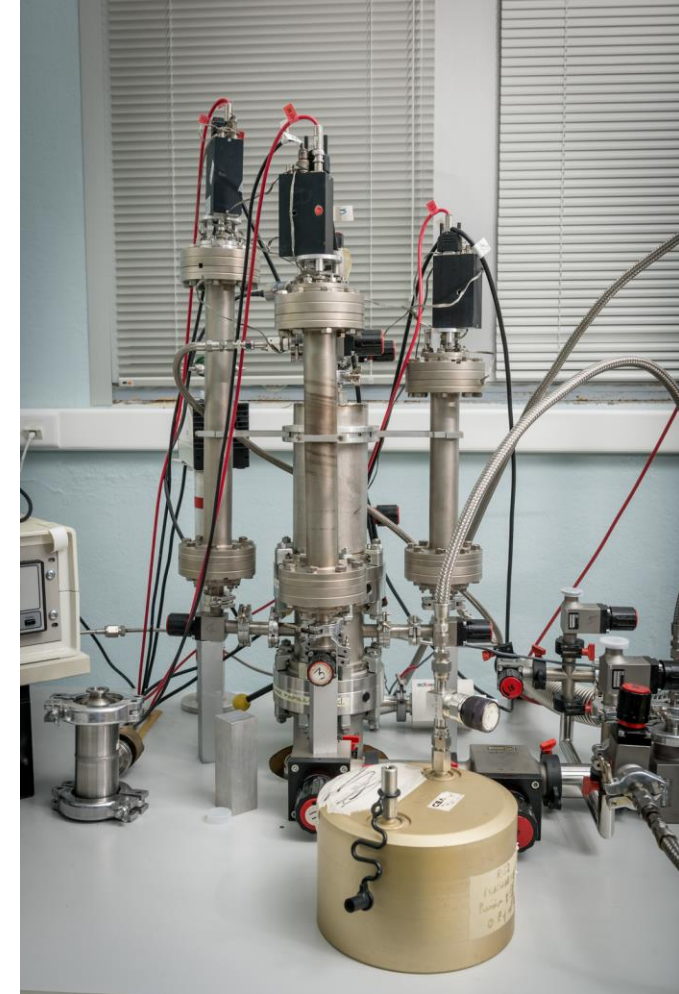
M. Unterweger, L. Johansson, L. Karam, M. Rodrigues and A. Yunoki.
Uncertainties in internal gas counting. Metrologia 52 (2015) S156-S164.

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Results:

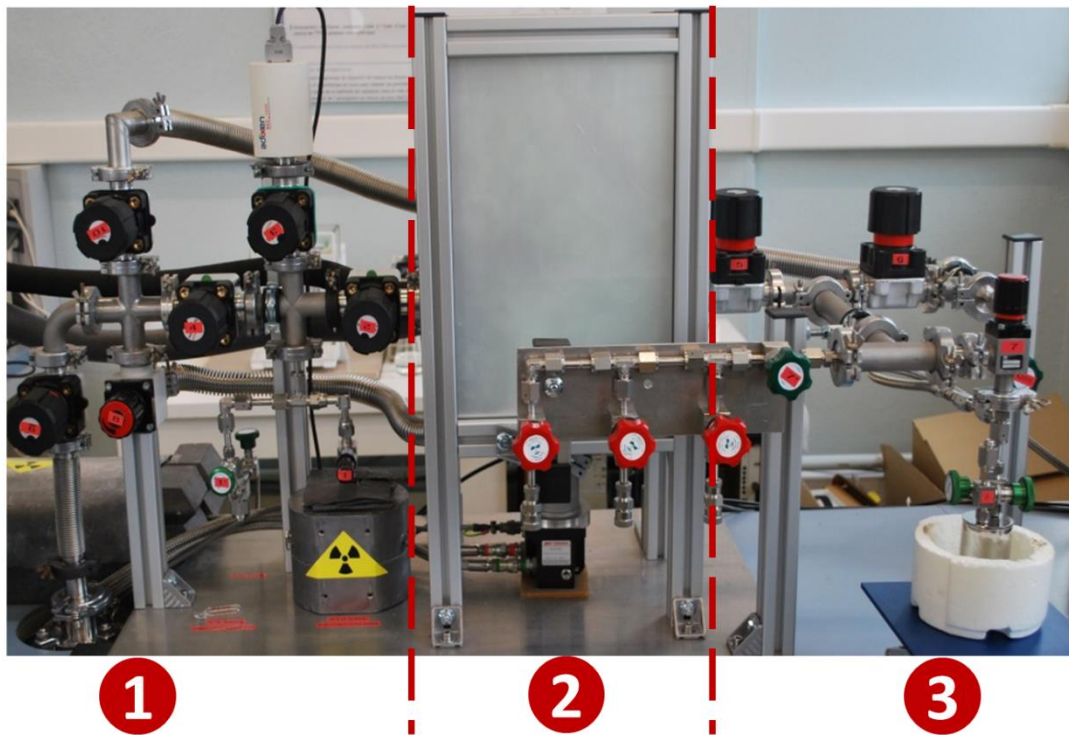
- ✓ Measurement of activity concentration (2.5 to 15 000 Bq·cm⁻³ at STP);
- ✓ Relative standard uncertainties:
 - ✓ 0.6% for ¹³³Xe and ⁸⁵Kr,
 - ✓ 1% for ¹²⁷Xe, ^{131m}Xe and ^{133m}Xe;
- ✓ Main uncertainties : counting statistics, dilution, volume, pressure;
- ✓ Validated by international comparisons:
 - ✓ BIPM.RI(II)–K1 for ¹³³Xe,
 - ✓ CCRI(II)–K2 for ⁸⁵Kr.



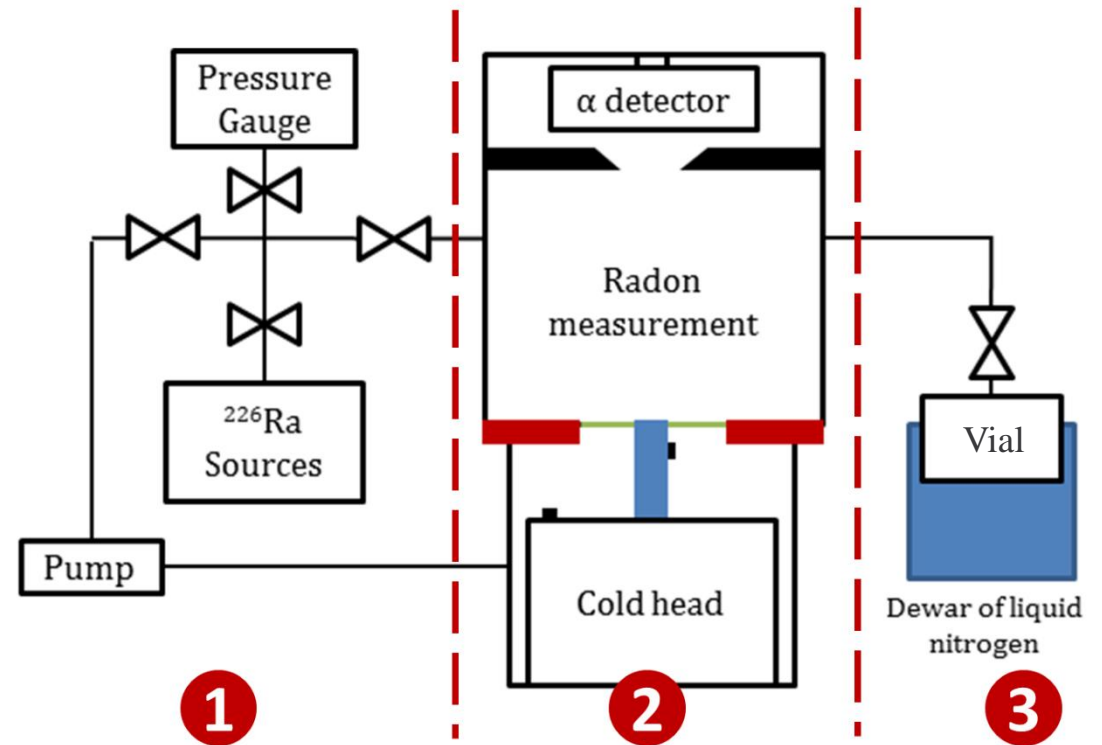
M. Unterweger, L. Johansson, L. Karam, M. Rodrigues and A. Yunoki.
Uncertainties in internal gas counting. Metrologia 52 (2015) S156-S164.

Alpha spectrometry on a frozen source of Rn-222. Method developed by J.L.Picolo (1996), upgraded in 2016.

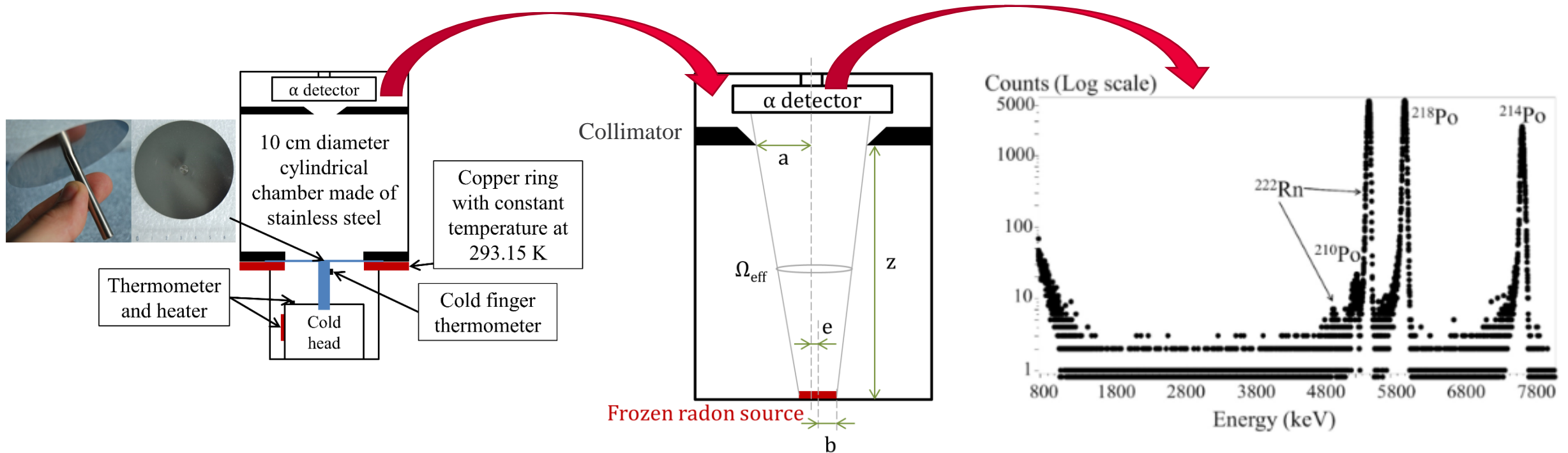
✓ ^{222}Rn is produced by a ^{226}Ra source and this gas is frozen below 100 K at 10^{-6} hPa.



Radon measurement system



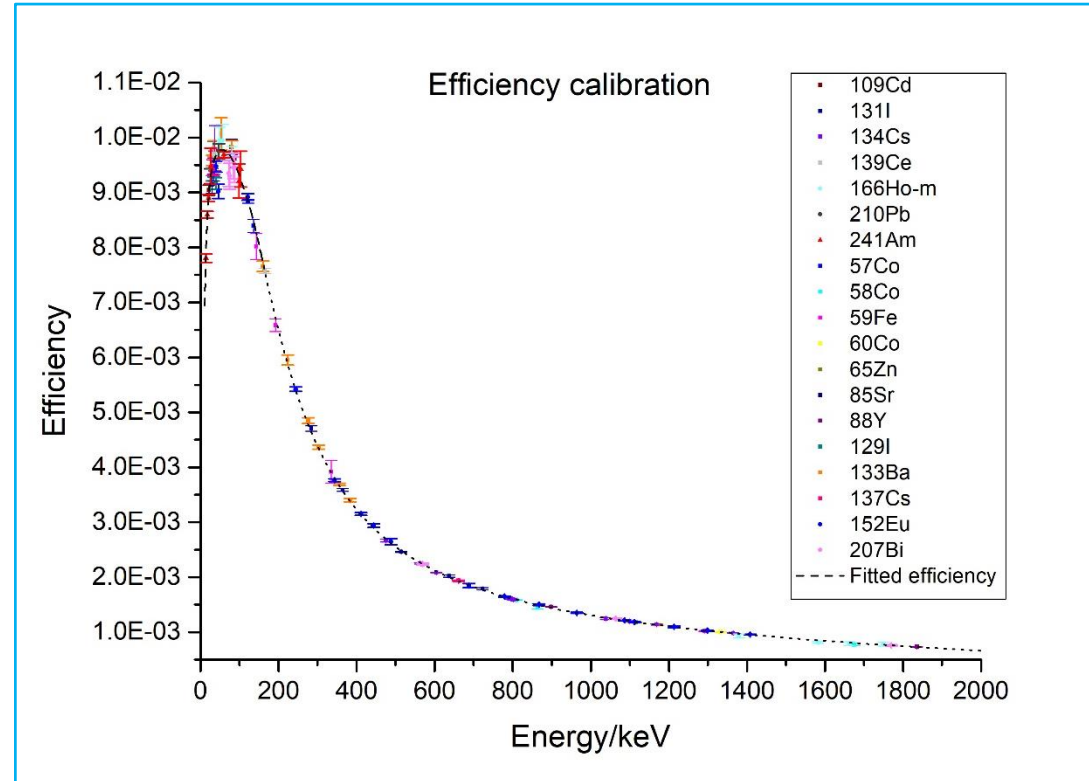
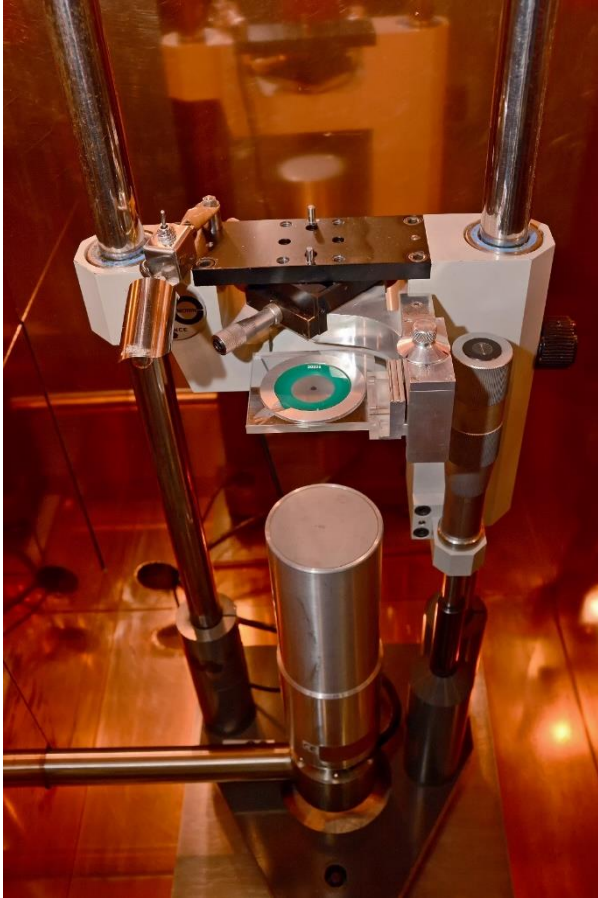
Simplified schematic representation



- ✓ Radon primary standard transferred in a metal vial;
- ✓ Measurement of ^{222}Rn activity from 100 Bq up to 4 MBq;
- ✓ Relative standard uncertainty: 0.4%;
- ✓ International comparison: CCRI(II)–K2.Rn-222.

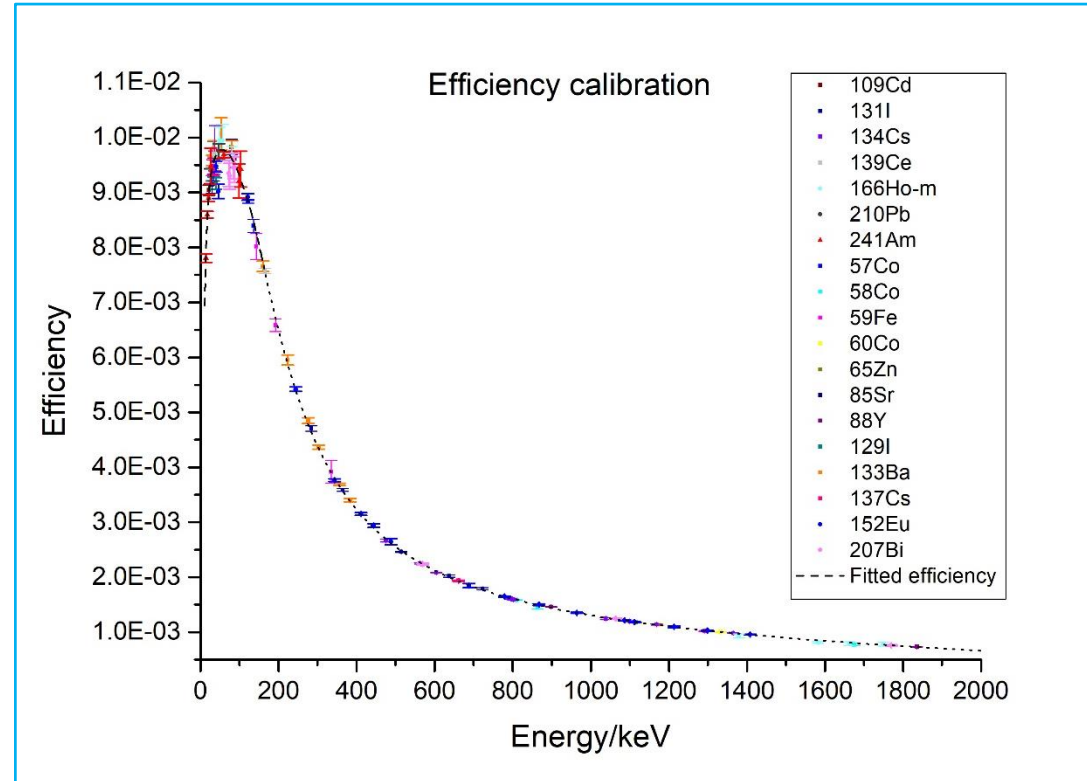
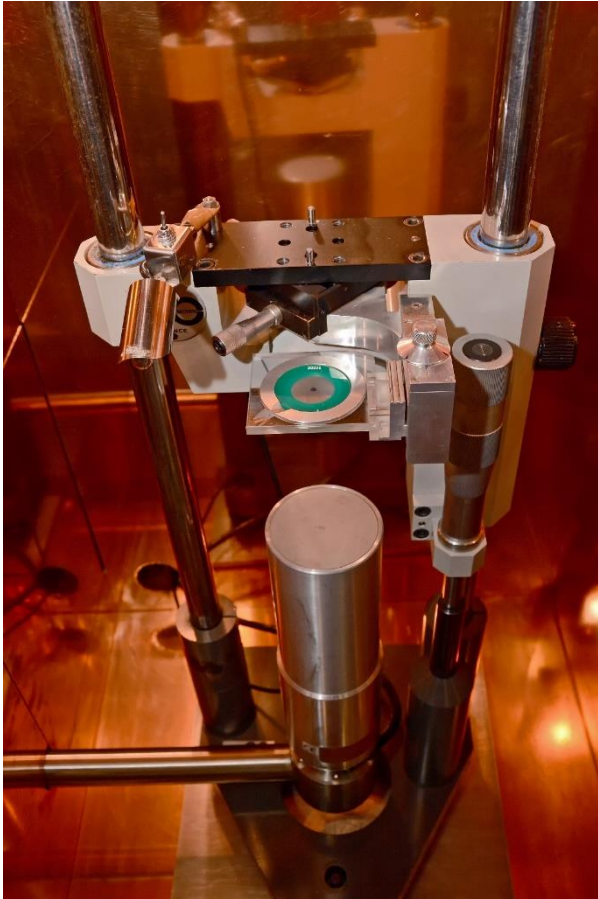
B. Sabot, S. Pierre, P. Cassette, *An absolute radon 222 activity measurement system at LNE-LNHB*, ARI 118 (2016) 167-174.

2 HPGe detectors. accurately calibrated for point sources at 10 cm from the detector window

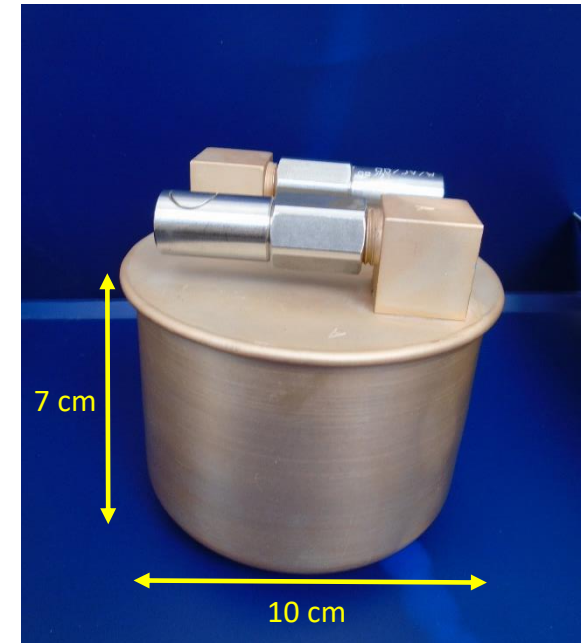


Relative uncertainty : 0.5% (high energy range)

2 HPGe detectors. accurately calibrated for point sources at 10 cm from the detector window



Relative uncertainty : 0.5% (high energy range)



Standardized geometry for
gas (500 cm³) (SG500)
Rounded bottom



2 measurement positions:

- ✓ "Contact"
- ✓ 8 cm from the detector window



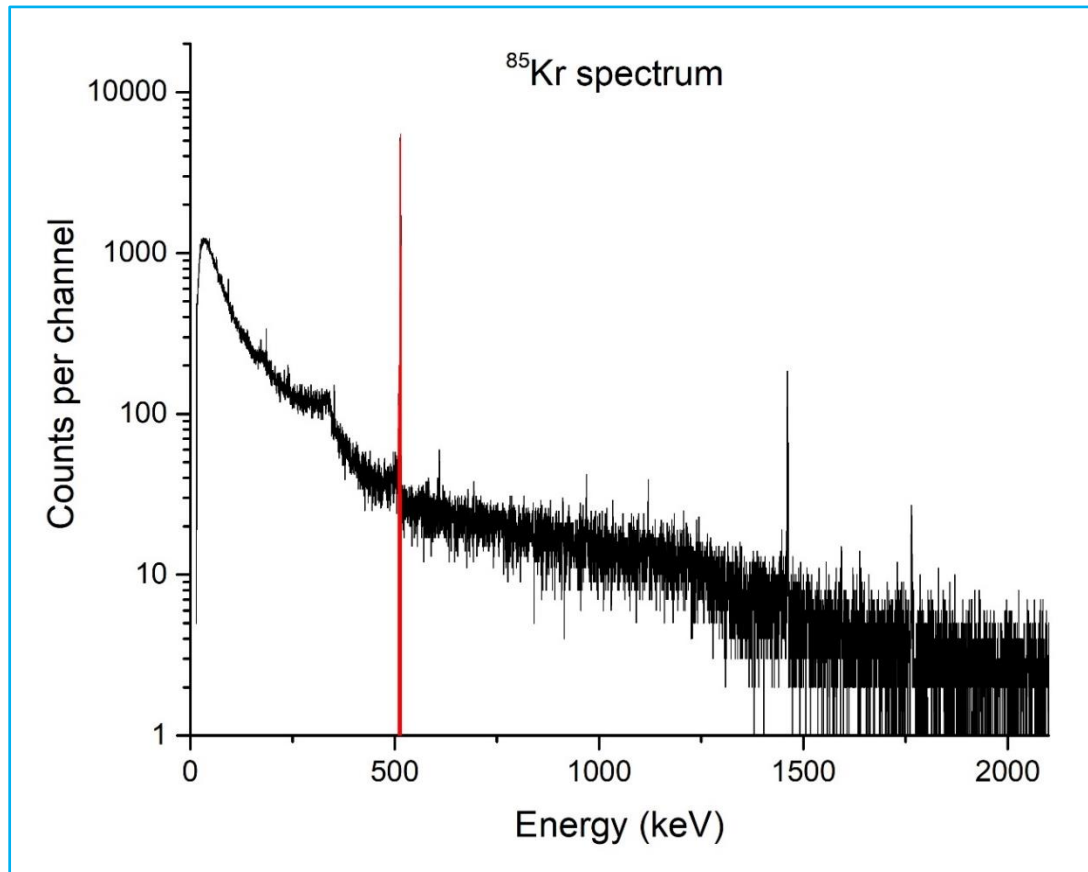
2 measurement positions:

- ✓ "Contact"
- ✓ 8 cm from the detector window

Dedicated 100 cm³ volume
(¹²⁷Xe and ²²²Rn)

3 reference containers measured on 2 HPGe detectors at 2 container-to-detector distances.

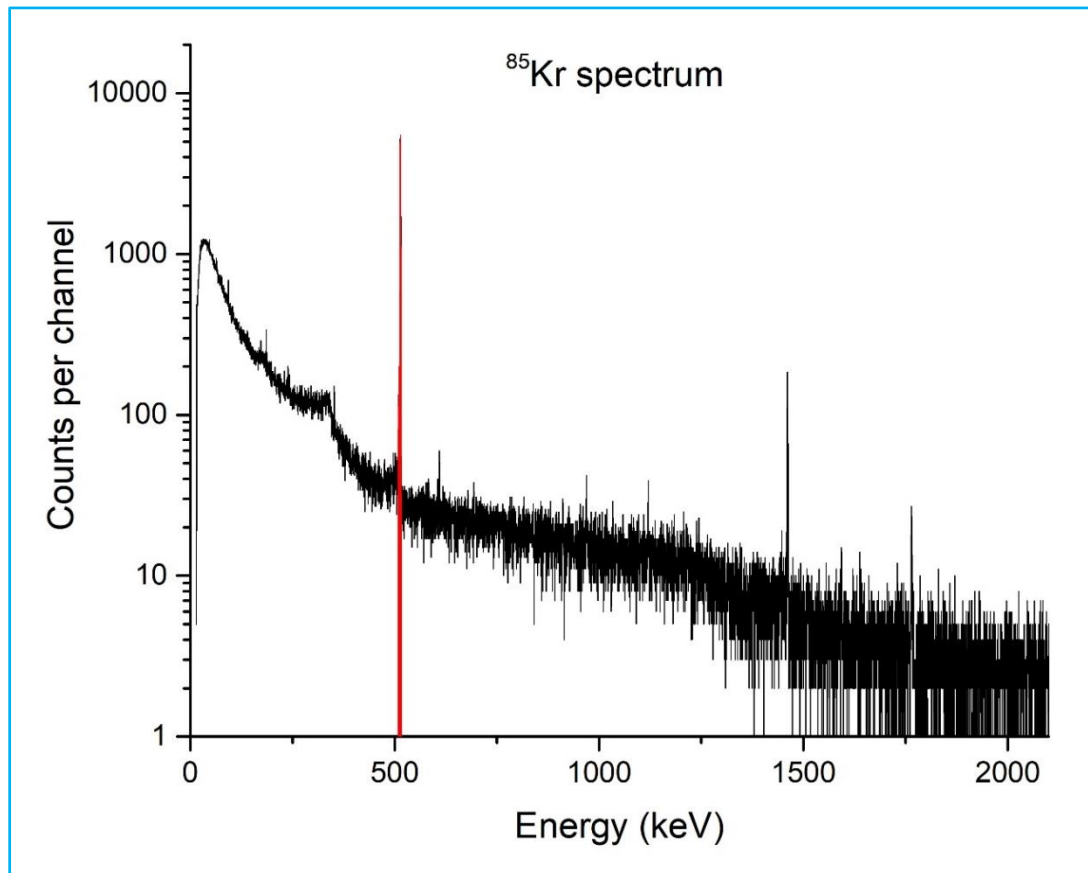
✓ The calibration factors are obtained from the count rate (N/t) recorded below the full-514-keV energy peak, reported to the reference date (C_T = Half-life correction) and corrected for decay during the measurement (C_{Dec}), divided the activity (33.49 (44) kBq).



$$CF = \frac{N \times C_T \times C_{Dec}}{t \times A}$$

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✓ The calibration factors are obtained from the count rate (N/t) recorded below the full-514-keV energy peak, reported to the reference date (C_T = Half-life correction) and corrected for decay during the measurement (C_{Dec}), divided the activity (33.49 (44) kBq).

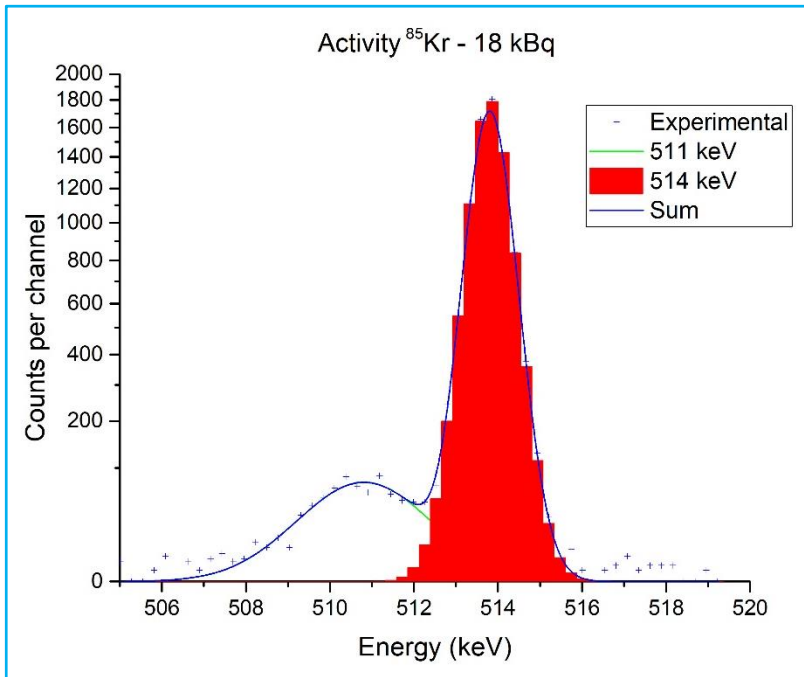


$$CF = \frac{N \times C_T \times C_{Dec}}{t \times A}$$

	Calibration factor for ⁸⁵ Kr (s ⁻¹ /Bq ⁻¹)	
Energy	514 keV	
Detector	Contact	8 cm
G8	3.60(5) 10 ⁻⁵	8.90(13) 10 ⁻⁶
G9	3.72(5) 10 ⁻⁵	9.22(13) 10 ⁻⁶

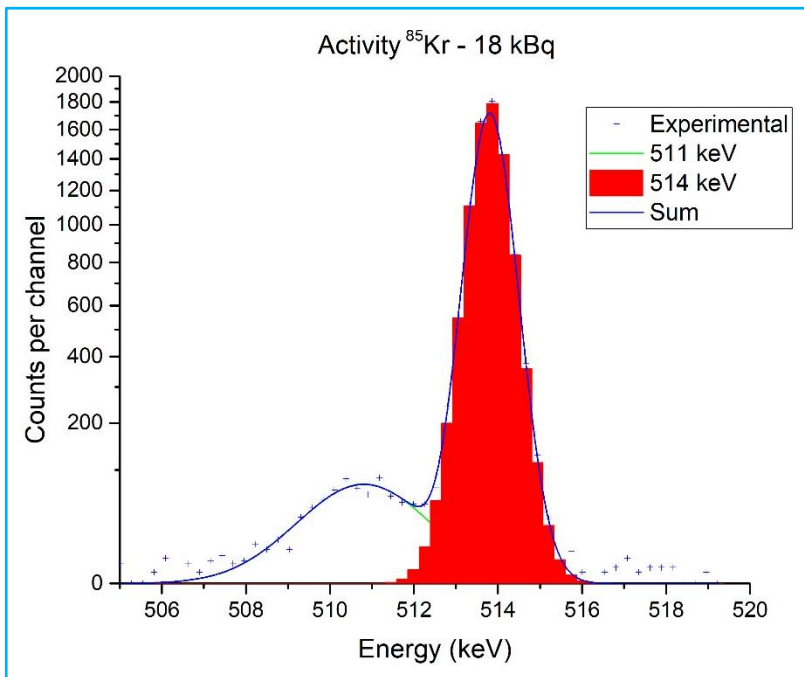
Main uncertainty : reference activity

Processing of the 511-514 keV region of interest

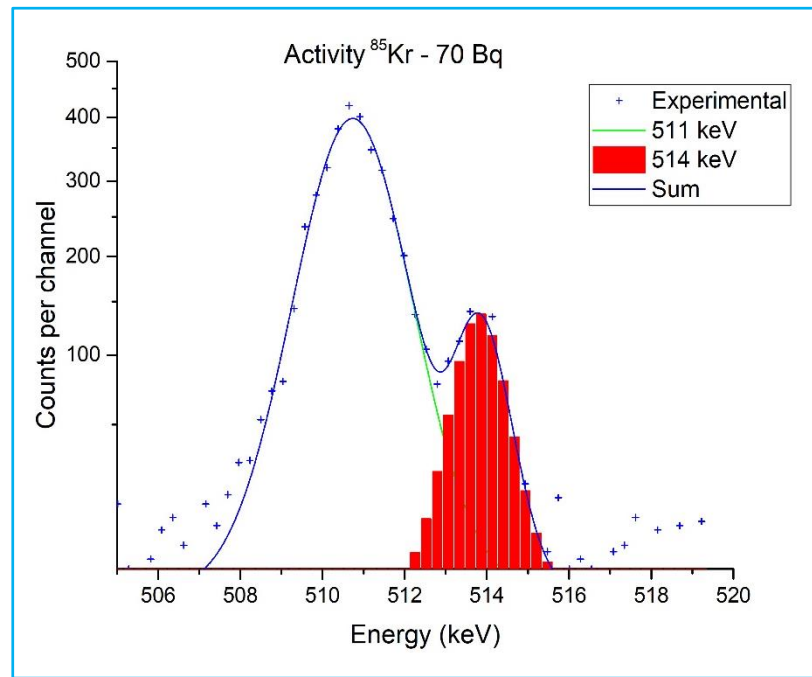


Activity ^{85}Kr = 18 kBq

Processing of the 511-514 keV region of interest



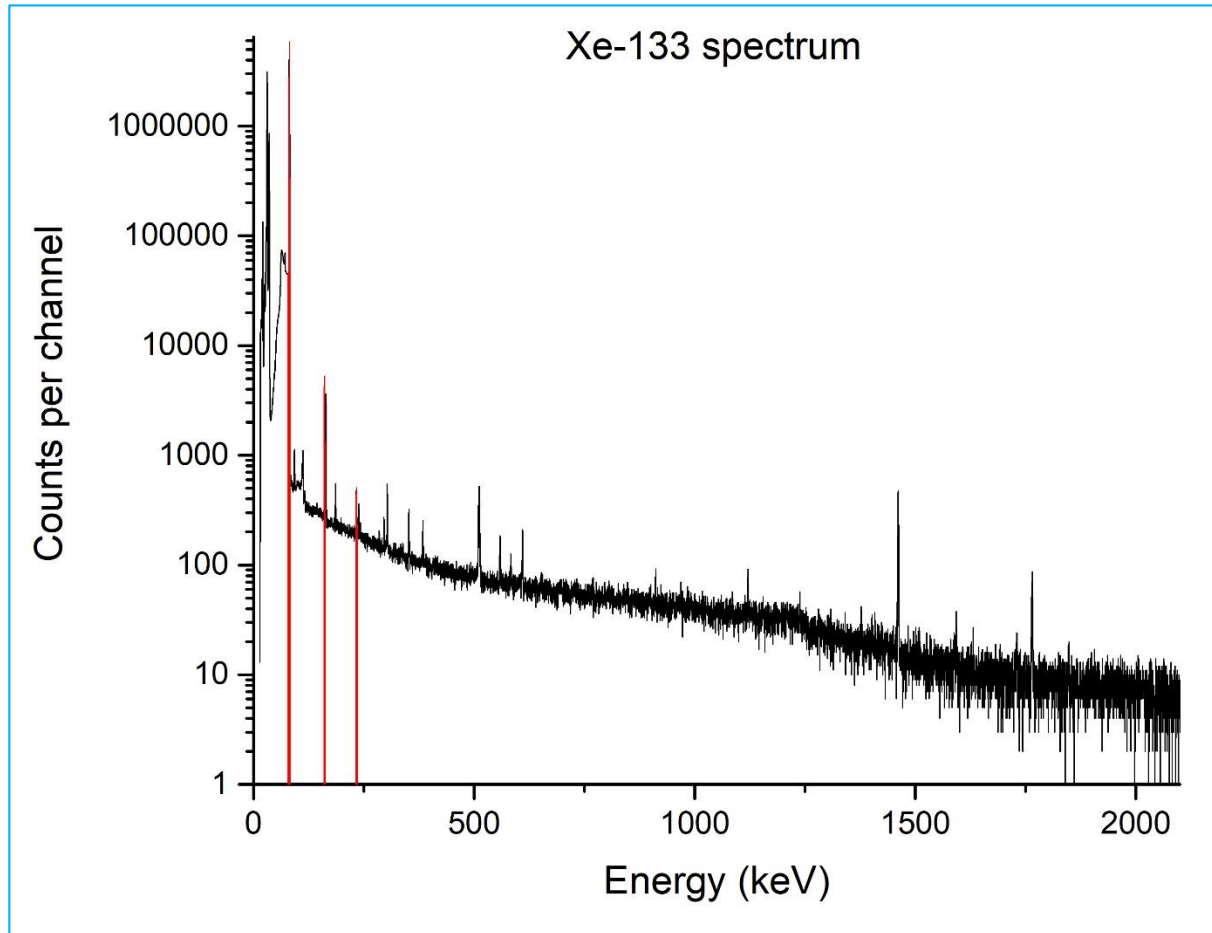
Activity ^{85}Kr = 18 kBq



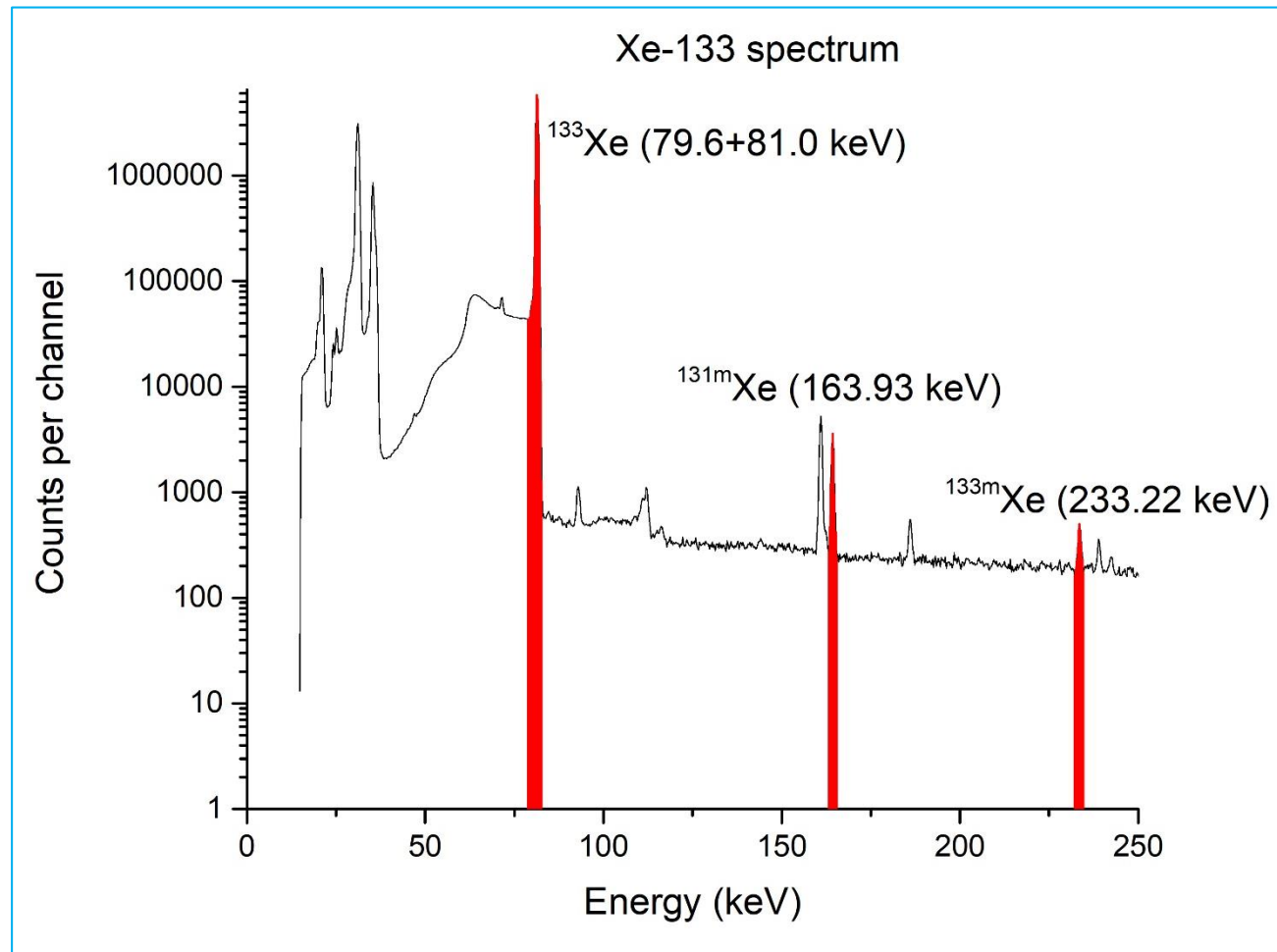
Activity ^{85}Kr = 70 Bq

- ✓ The width of the annihilation peak (511 keV) must be taken into account when determining the area of the peak of interest at 514 keV.
- ✓ Use the energy resolution calibration for accurate fitting (e.g. COLEGRAM code)!
- ✓ Gaussian standard deviation (σ):
 - ✓ 511 keV : 1.01 keV
 - ✓ 514 keV: 0.49 keV

Quantification of the impurities → One container directly measured for $2 \cdot 10^5$ s



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$$A(\text{Bq}) = \frac{N(E) \times C_T \times C_{Dec}}{t \times I_G(E) \times \varepsilon(E) \times TE}$$

Count rate $(N(E)/t)$ in the full energy peak,

C_T = Half-life correction,

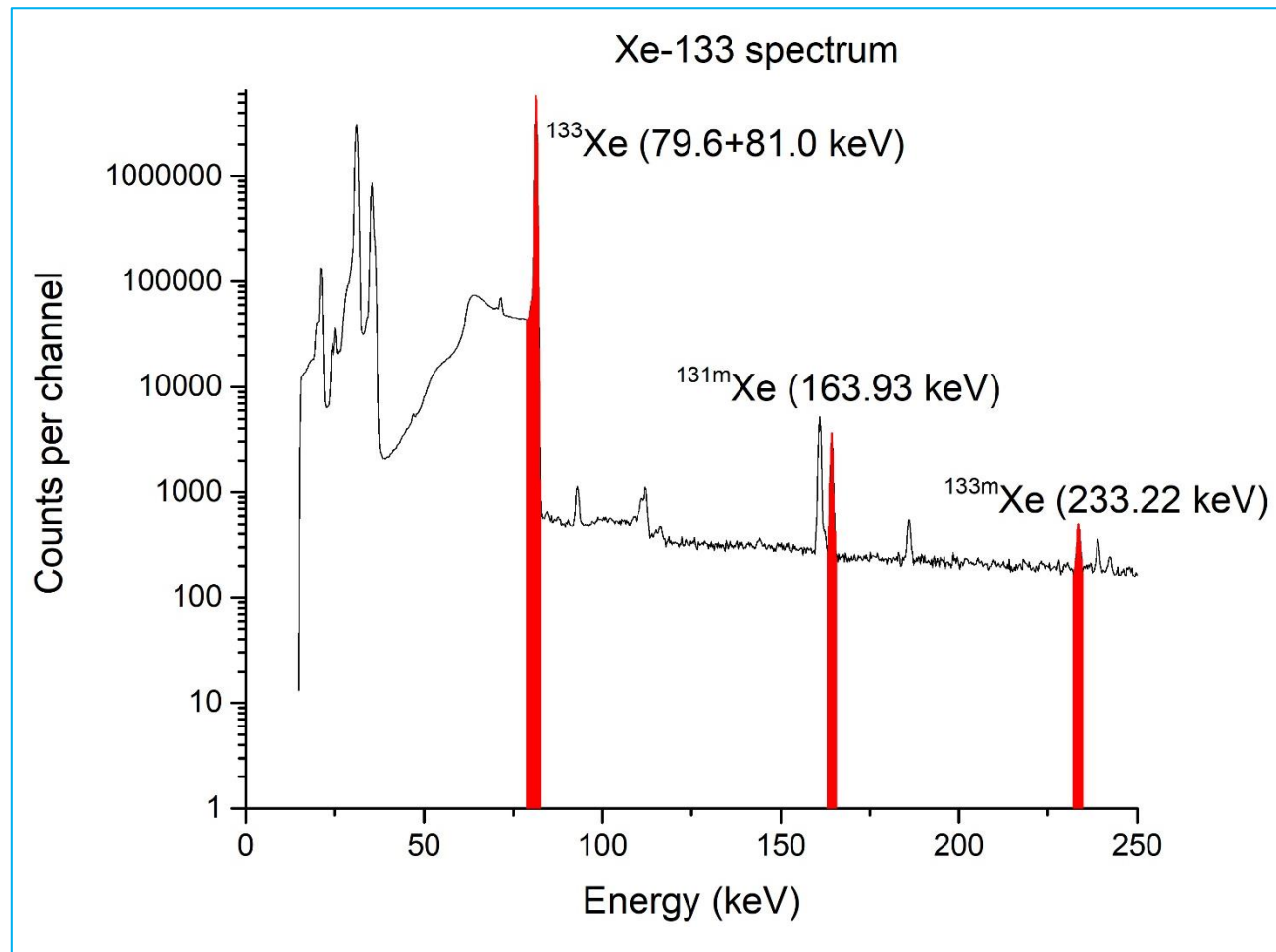
C_{Dec} = Correction for decay during the measurement,

$I_G(E)$ = photon emission intensity,

$\varepsilon(E)$ = full-energy peak efficiency (point source),

TE = efficiency transfer factor.

Quantification of the impurities → One container directly measured for $2 \cdot 10^5$ s



$$A(\text{Bq}) = \frac{N(E) \times C_T \times C_{Dec}}{t \times I_G(E) \times \varepsilon(E) \times TE}$$

- ✓ Efficiency transfer (TE from ETNA code) use to get the activities;
- ✓ $^{131\text{m}}\text{Xe}$: 118 Bq,
- ✓ $^{133\text{m}}\text{Xe}$: 7.4 Bq;
- ✓ ^{133}Xe : 10.3 (5) kBq;
- ✓ In relative proportions:
1.14% Bq/Bq in $^{131\text{m}}\text{Xe}$,
0.07% Bq/Bq in $^{133\text{m}}\text{Xe}$.

Two steps

- ✓ Quantification of the impurities → correction factors for the absolute measurement of activity,
- ✓ 10 reference SG500 containers.

At the reference date, the ^{133}Xe activity in each container was 10.72 (11) kBq.

The calibration factors are obtained from the count rate (N/t) recorded below the 79-81 keV doublet.

$$CF = \frac{N \times C_T \times C_{Dec}}{t \times A}$$

	Calibration factor for ^{133}Xe ($\text{s}^{-1}/\text{Bq}^{-1}$)	
Energy	79-81 keV	
Detector	Contact	8 cm
G8	$1.229(14) \cdot 10^{-2}$	$2.826 (32) \cdot 10^{-3}$
G9	$1.202(13) \cdot 10^{-2}$	$2.743(46) \cdot 10^{-3}$

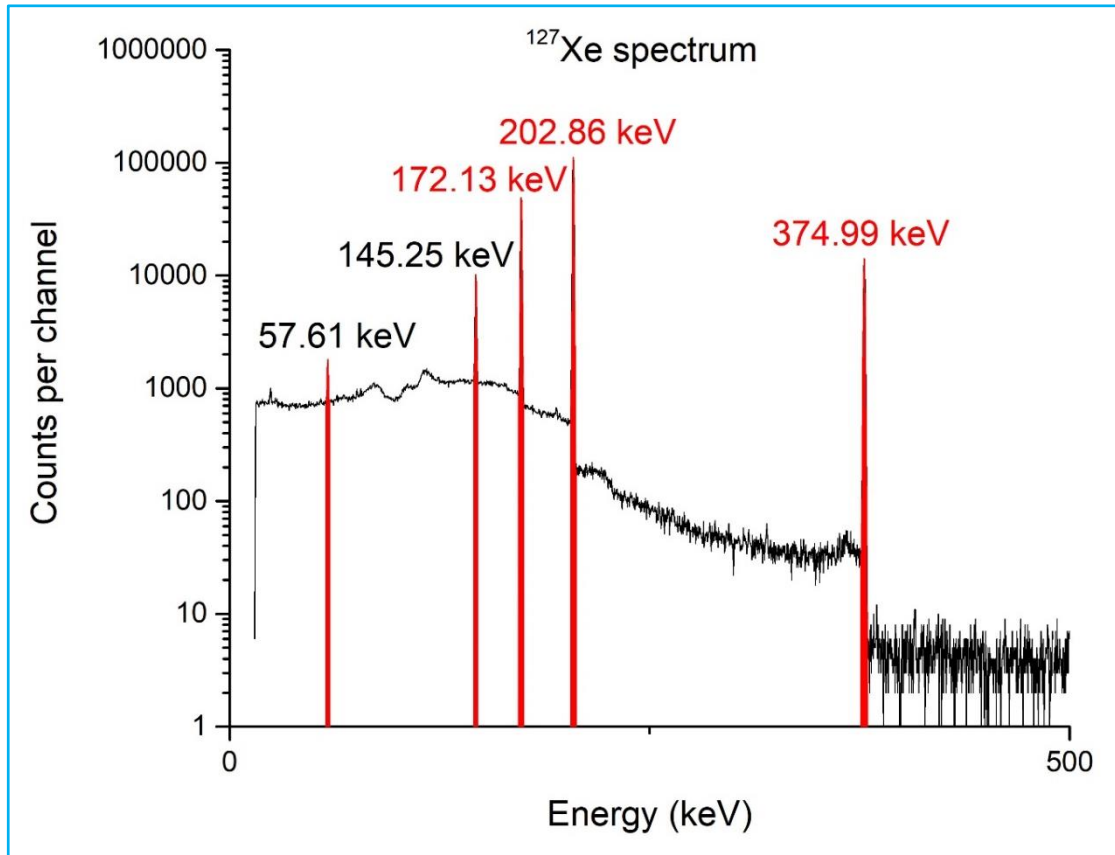
Main uncertainty : reference activity

- ✓ Comparison between experiment and calculation (efficiency transfer + photon emission intensity),
- ✓ Ratio of the calibration factors (calculation/experiment):

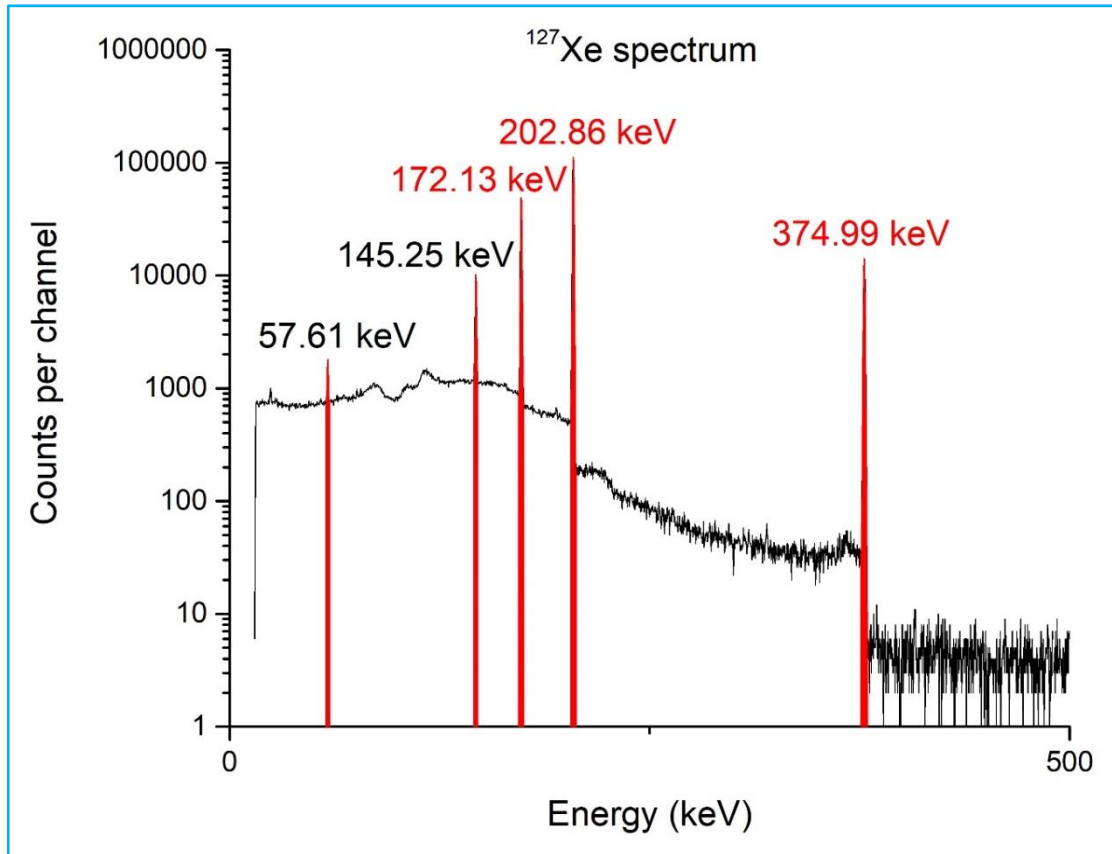
	Calibration factor (s^{-1}/Bq^{-1})	
Radionuclide	^{131m}Xe	^{133m}Xe
Energy	164 keV	232 keV
Detector G8 contact	$5.16 (27) 10^{-4}$	$1.98 (10) 10^{-3}$

	^{85}Kr (E = 514 keV – 0.435%)		^{133}Xe (E = 79.61 + 81.00 keV – 37.28%)	
Measurement distance	Contact	8 cm	Contact	8 cm
G8	1.056	1.011	1.067	1.025
G9	1.037	1.016	1.021	1.008





Reference activity (triple counters)
Efficiency = 0.967(4)



Reference activity (triple counters)
Efficiency = 0.967(4)

Use of the 100 cm³ container;
 ✓ Distance container-detector = 6.83 cm
 ✓ Reference activity : 12.58 (14) kBq

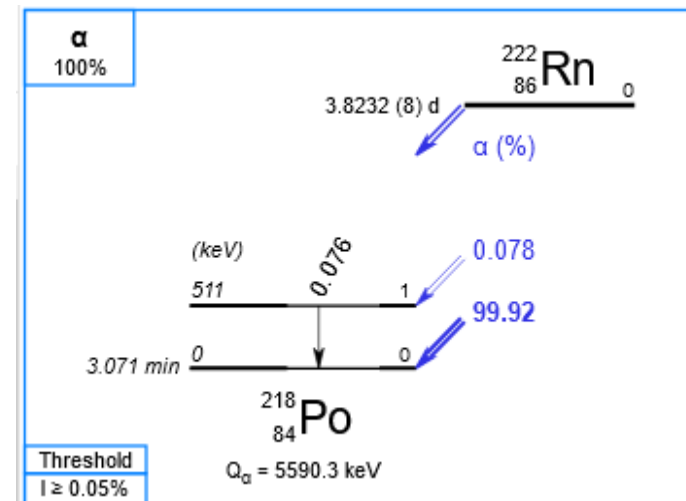
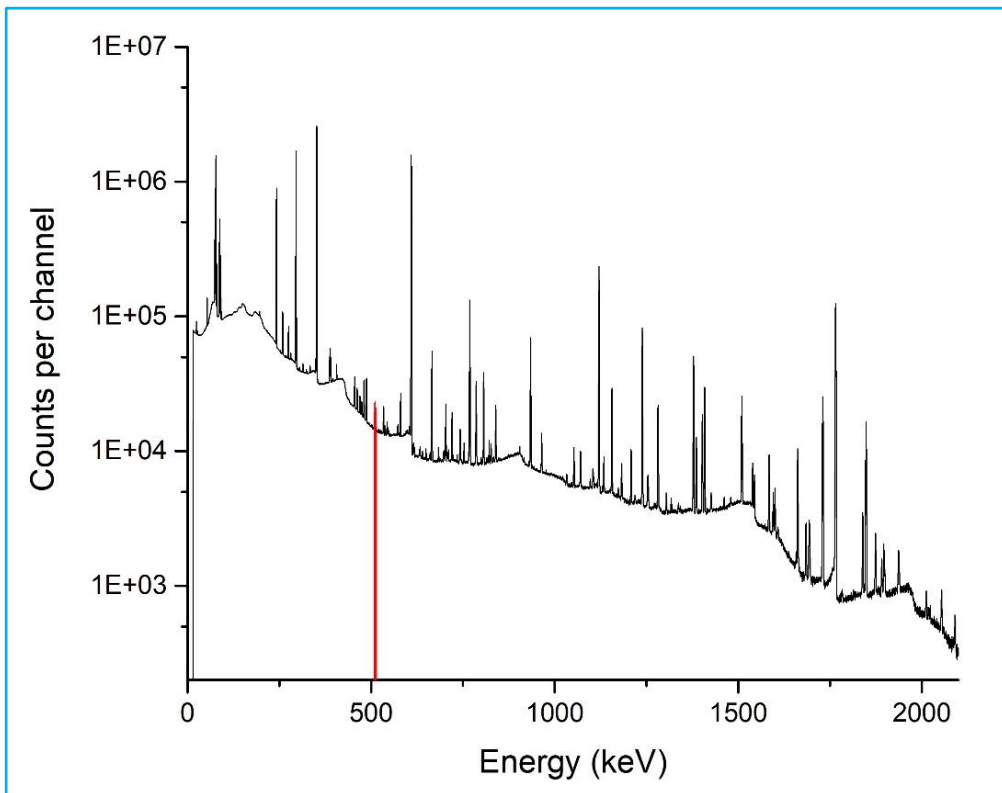
3 main peaks

✓ 172 keV, 202 keV and 375 keV

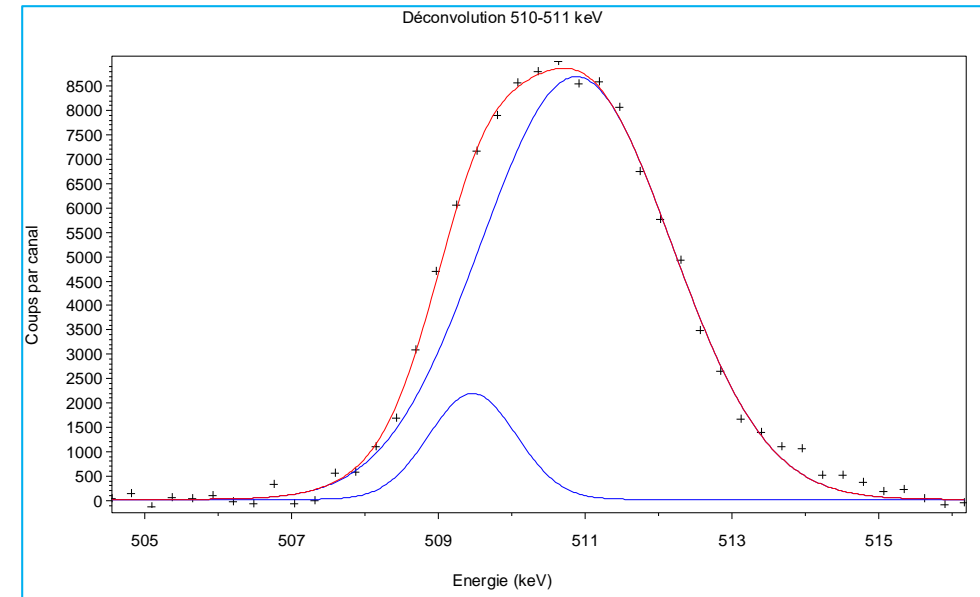
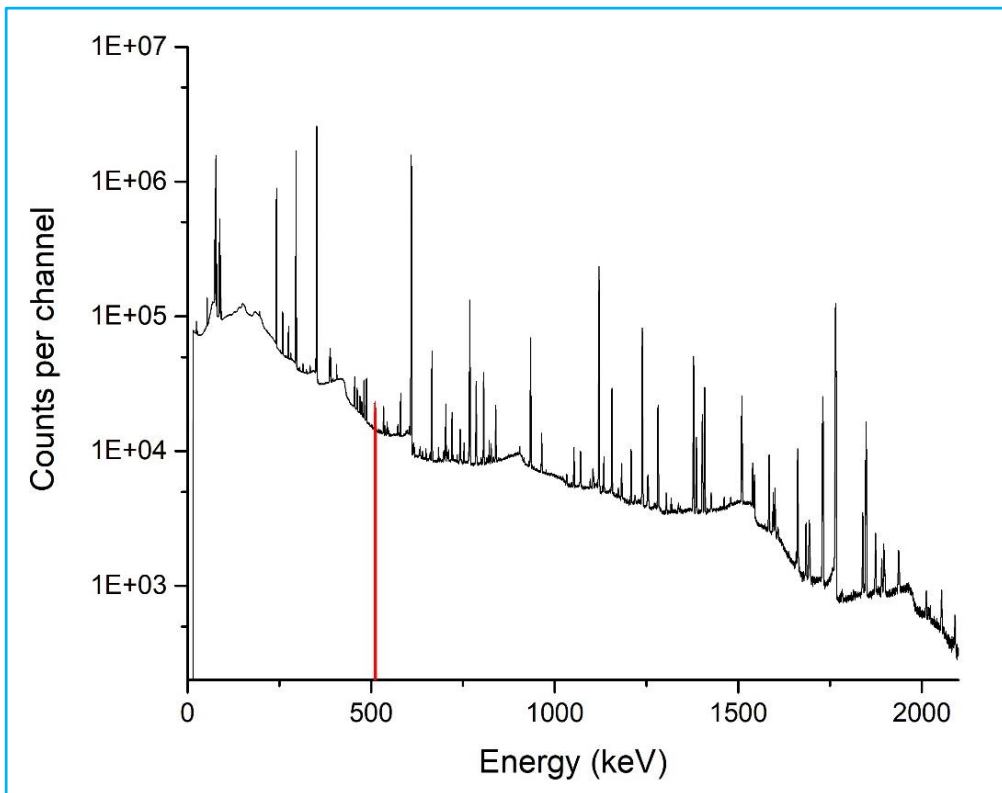


	Calibration factors for ¹²⁷ Xe (s ⁻¹ /Bq ⁻¹)		
Energy	172 keV	203 keV	375 keV
G8	1.91 (2) 10 ⁻³	4.62 (5) 10 ⁻³	6.83 (8) 10 ⁻⁴
G9	1.86 (2) 10 ⁻³	4.53 (5) 10 ⁻³	6.95 (8) 10 ⁻⁴

- ✓ Reference activity: alpha spectrometry on frozen source : 39.44 (13) kBq;
- ✓ Difficult to directly measure the calibration factor → large uncertainty;
- ✓ Use of the 100 cm³ container – Acquisition for 300 000 s.



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- ✓ Use of the 100 cm³ container – Acquisition for 300 000 s.



Only 10% compared to annihilation peak
Gaussian width (σ): 0.59 and 1.30 keV

Energy	Calibration factor for Rn-222 (510 keV)
G8	$1.62(8) \cdot 10^{-6}$

New practical tool to calculate activities of radionuclides in a decay chain

<http://www.lnhb.fr/donnees-nucleaires/module-lara/>

Hide/show main panel

Nuclide list:

218At
218Rn
219At
219Rn
220Rn
221Fr
221Ra
222Rn

Nuclide, element or mass number search:
or
(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

Emissions: X Gamma Alpha

Language: EN EO FR

Nuclide search criteria

Decay mode: β^+ , ϵ β^- IT α
(And Or XOR)

Emissions: X Gamma Alpha

Energy 1 (or range): \pm / - keV

Energy 2 (or range): \pm / - keV } And

Energy 3 (or range): \pm / - keV } Or

Intensity range: - %

Mass range: - u

Atomic number range: -

Half-life range: a - a

UTC date & time: 2022-04-21 - 08:45:38

Last update: 2021-09-21 (data) & 2022-04-15 (code)

- Quick user's guide
- Printed version (2015) available by [EDP Sciences](#)
- [Contact us](#) (suggestions, comments, remarks, ...)
- Back to [LNHB home page](#)

1 490 (20)	2 (1)	y	Pb-210	-1	6	Tl-210
1 540 (30)	2 (1)	y	Pb-210	-1	7	Tl-210
1 590 (30)	2 (1)	y	Pb-210	8	4	Tl-210
1 650 (30)	2 (1)	y	Pb-210	-1	8	Tl-210
83 (30)	1.98 (40)	y	Pb-210	-1	1	Tl-210
46.539 (1)	4.252 (40)	y	Bi-210	1	0	Pb-210
304.896 (6)	26 (5)	y	Tl-206	2	0	Hg-206
649.42 (5)	2.2 (3)	y	Tl-206	3	0	Hg-206
344.52 (17)	0.55 (11)	y	Tl-206	3	2	Hg-206
5 304.33 (7)	99.99876 (4)	α	Po-210	0	0	Po-210

Tools

Activity \rightleftharpoons Mass conversion: Bq \rightleftharpoons g

Decay calculation: calculation step(s)

$t_1 \rightarrow t_2 = 0 \rightarrow 3.823E0$ d

$d_1 \rightarrow d_2 = 04 \ 21 \ 2022 \ 10:44:36 \rightarrow 04 \ 22 \ 2022 \ 10:44:36$

Nuclide	(T _{1/2})	A ₀	A(t ₂)	A*(t ₂)	A _T (t ₂)	A _T /A _{parent}
222Rn	(3.8232 d)	1000	5.000E+2		5.000E+2	Bq 1
218Po	(3.071 min)	0	0.000E+0	5.003E+2	5.003E+2	Bq 1.001E+0
218At	(1.4 s)	0	0.000E+0	1.101E-1	1.101E-1	Bq 2.202E-4
214Pb	(26.916 min)	0	0.000E+0	5.026E+2	5.026E+2	Bq 1.005E+0
218Rn	(36.0 10 ⁻³ s)	0	0.000E+0	1.101E-4	1.101E-4	Bq 2.202E-7
214Bi	(19.8 min)	0	0.000E+0	5.046E+2	5.046E+2	Bq 1.009E+0
214Po	(162.3 10 ⁻⁶ s)	0	0.000E+0	5.045E+2	5.045E+2	Bq 1.009E+0
210Tl	(1.30 min)	0	0.000E+0	1.060E-1	1.060E-1	Bq 2.120E-4
210Pb	(22.23 a)	0	0.000E+0	2.332E-1	2.332E-1	Bq 4.664E-4
210Bi	(5.012 d)	0	0.000E+0	5.683E-2	5.683E-2	Bq 1.137E-4
206Hg	(8.32 min)	0	0.000E+0	4.425E-9	4.425E-9	Bq 8.850E-12
210Po	(138.3763 d)	0	0.000E+0	3.934E-4	3.934E-4	Bq 7.868E-7
206Tl	(4.202 min)	0	0.000E+0	8.384E-8	8.384E-8	Bq 1.677E-10
206Pb	(stable)					
Total						2.513E+3 Bq 5.026E+0

(d₂ - d₁ = d)

Scheme

Threshold $I \geq 0.05\%$
Q_α = 5560.3 keV

New practical tool to calculate activities of radionuclides in a decay chain

<http://www.lnhb.fr/donnees-nucleaires/module-lara/>

Tools

Activity \rightleftharpoons Mass conversion: Bq \rightleftharpoons g

Decay calculation: calculation step(s)

: $t_1 \rightarrow t_2 =$ \rightarrow

: $d_1 \rightarrow d_2 =$ \rightarrow

Nuclide	(T½)	A ₀	A(t ₂)	A*(t ₂)	A _T (t ₂)	Bq	A _T /A _{parent}
²²² Rn	(3.8232 d)	<input type="text" value="1000"/>	<input type="text" value="5.000E+2"/>		<input type="text" value="5.000E+2"/>	Bq	<input type="text" value="1"/>
²¹⁸ Po	(3.071 min)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="5.003E+2"/>	<input type="text" value="5.003E+2"/>	Bq	<input type="text" value="1.001E+0"/>
²¹⁸ At	(1.4 s)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="1.101E-1"/>	<input type="text" value="1.101E-1"/>	Bq	<input type="text" value="2.202E-4"/>
²¹⁴ Pb	(26.916 min)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="5.026E+2"/>	<input type="text" value="5.026E+2"/>	Bq	<input type="text" value="1.005E+0"/>
²¹⁸ Rn	(36.0 10 ⁻³ s)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="1.101E-4"/>	<input type="text" value="1.101E-4"/>	Bq	<input type="text" value="2.202E-7"/>
²¹⁴ Bi	(19.8 min)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="5.046E+2"/>	<input type="text" value="5.046E+2"/>	Bq	<input type="text" value="1.009E+0"/>
²¹⁴ Po	(162.3 10 ⁻⁶ s)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="5.045E+2"/>	<input type="text" value="5.045E+2"/>	Bq	<input type="text" value="1.009E+0"/>
²¹⁰ Tl	(1.30 min)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="1.060E-1"/>	<input type="text" value="1.060E-1"/>	Bq	<input type="text" value="2.120E-4"/>
²¹⁰ Pb	(22.23 a)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="2.332E-1"/>	<input type="text" value="2.332E-1"/>	Bq	<input type="text" value="4.664E-4"/>
²¹⁰ Bi	(5.012 d)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="5.683E-2"/>	<input type="text" value="5.683E-2"/>	Bq	<input type="text" value="1.137E-4"/>
²⁰⁶ Hg	(8.32 min)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="4.425E-9"/>	<input type="text" value="4.425E-9"/>	Bq	<input type="text" value="8.850E-12"/>
²¹⁰ Po	(138.3763 d)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="3.934E-4"/>	<input type="text" value="3.934E-4"/>	Bq	<input type="text" value="7.868E-7"/>
²⁰⁶ Tl	(4.202 min)	<input type="text" value="0"/>	<input type="text" value="0.000E+0"/>	<input type="text" value="8.384E-8"/>	<input type="text" value="8.384E-8"/>	Bq	<input type="text" value="1.677E-10"/>
²⁰⁶ Pb	(stable)						
Total					<input type="text" value="2.513E+3"/>	Bq	<input type="text" value="5.026E+0"/>

(d₂ - d₁ = d)

[Copy table to clipboard](#)

- ✓ Calibration factors for two detectors for gas geometries;
- ✓ Direct activity measurement for ^{85}Kr and ^{133}Xe for standard 500 cm³ containers (relative uncertainty about 1.2%);
 - ✓ Possibility to reduce annihilation contribution (anti-cosmic veto);
 - ✓ Calibration factors established for ^{133}Xe impurities ($^{131\text{m}}\text{Xe}$ and $^{133\text{m}}\text{Xe}$) (relative uncertainty about 5%);
- ✓ Calibration factor for ^{127}Xe for 100 cm³ container (relative uncertainty about 1.1%);
- ✓ More difficulties for ^{222}Rn : complementary measurements on-going (one experiment (1956) for the photon emission !).

PHYSICAL REVIEW

VOLUME 102, NUMBER 2

APRIL 15, 1956

Decay of Rn^{220} and $\text{Rn}^{222}\dagger$

L. MADANSKY AND F. RASETTI
Johns Hopkins University, Baltimore, Maryland
(Received January 17, 1956)

A gamma ray of energy 510 ± 2 kev and approximate intensity 7×10^{-4} quanta per decay is emitted in the alpha decay of Rn^{222} . A gamma ray of energy 542 ± 2 kev and approximate intensity 2.5×10^{-4} quanta per decay is emitted in the alpha decay of Rn^{220} . The intensities are considered accurate only to within a factor of two.



Thank you for your attention !