



# Decay data measurements using MMCs at CEA-LNHB

#### **Matias Rodrigues**

Martin Loidl Arshjot Kaur (PhD student) Mostafa Zahir (PhD student)

MiND Workshop, June 28, 2023



# Outline

- Introduction
  - Presentation of the CEA-LNHB
  - Short history of the MMCs at LNHB
- Decay data by decay energy spectrometry
  - Beta shape of <sup>63</sup>Ni and <sup>241</sup>Pu
  - BR and end-point energy of <sup>151</sup>Sm
  - Electron capture probabilities of <sup>55</sup>Fe
- Absolute L X-ray emission intensities of actinides
  - The MMC SMX3
  - L X-rays from the decay of <sup>238</sup>Pu, <sup>244</sup>Cm <sup>233</sup>Pa, <sup>237</sup>Np
  - L X-rays from the decay of <sup>241</sup>Am

# Introduction

# The Laboratoire National Henri Becquerel Activity Metrology



#### Conventional methods for activity standardization

- 4πβ-γ coincidence
- α-particle counting by defined solid angle
- Liquid scintillation (TDCR method)
- Triple internal gas counting
- And others...

#### Decay data measurements and evaluations

- Beta and gamma-ray spectrometry
- Monochromatic sources of photons
- Code Beta shape
- DDEP Decay Data Evaluation Project (Data recommendation dissemination of data http://www.lnhb.fr/donnees-nucleaires/module-lara/)
- Metallic magnetic calorimeters

#### Radioactive source preparation

- Authorizations to prepare and to measure many radionuclides with large activity,
- Liquid, solid or gaseous radioactive source
- Radiochemical separation
- Source activities with low uncertainties

#### MMC at LNHB: started in 2004 with the direct coupling



- Electroprecipited <sup>241</sup>Pu source (8 Bq) between two 12 µm thick gold foils
- Energy calibration: external <sup>55</sup>Fe source
- Data recorded continuously 2.6 days @ 16 mK



First measurement of the beta spectrum of <sup>241</sup>Pu with a cryogenic detector

M. Loidl \*, M. Rodrigues, B. Censier, S. Kowalski, X. Mougeot, P. Cassette, T. Branger, D. Lacour



#### MMC at LNHB : 2008, Coupling with meander shape pick-up coil





# MMC chips produced in collaboration with KIP 🔷 🎲 Heidelberg 2010-2021 and KIT Karlsruhe 2021-now



For a given radionuclide and a given spectrometry, absorber sized is to achieve the required efficiency.

- $\rightarrow$  The sensor MMC size chosen such as the sensor matches the absorber heat capacity
- $\rightarrow$  The source+absorber prepared at LNHB and attached to the sensor by gluing

# **Decay data by decay** energy spectrometry

# Beta spectrum shapes of <sup>241</sup>Pu and <sup>63</sup>Ni



# Impact on the activity measurement by LSC

**E** =

Counting efficiency in liquid scintillation depends on the beta spectrum for the 2 LSC methods:

 $(S(E)(1-e^{-\eta})^2 dE)$ 

- CIEMAST/NIST (CN) (2 PMTs)
- TDCR (triple to double coincidence ratio) (3 PMTs) TDCR =





→Better agreement between the 2 LSC methods taking into account the exchange effect

 $(S(E)(1-e^{-\eta})^3 dE)$ 



Recommended value DDEP: 99.07 (4)% and 0.93 (4)%

• Measured  $E_{max} = 76.430$  (68) keV.

Recommended value AME2020: 76.5 (5) keV

Beta spectrum of <sup>151</sup>Sm





K. Kossert, ARI, Vol. 185, 110237, 2022 https://doi.org/10.1016/j.apradiso.2022.110237

# **Electron capture probabilities (EC) of 55Fe**

Like pure beta emitters to the GS, EC radionuclides to the GS can only be measured in LSC Absorber with <sup>55</sup>Fe

10<sup>4</sup>

- $\rightarrow$  EC probabilities is required.
- Source prepared by electroplating on Au foil
- Source foil with ~ 10 Bq of  ${}^{55}$ Fe between two Au foils

L Captures

10<sup>3</sup>

(770 eV)

Energy [eV]

Absorber dimensions 600 µm × 600 µm × 24 µm

1 week data taking i.e.  $4 \times 10^6$  counts



M. Loidl, et al. ARI, Vol. 134, P395, 2018 https://doi.org/10.1016/j.apradiso.2017.10.042

**M** Captures

10<sup>2</sup>

(84 eV)



1200 r

1000

800

Counts

400

# 

# "Absolute" photon emission intensities (PEIs)

- Absolute PEIs: *I* = number of photons at *E* per 100 decays
- Essential decay parameter for quantitative analysis by photon spectrometry
- Absolute PEIs are challenging to measure accurately with standard deviation < 1%...



•  $\varepsilon_{FEP}(E)$  is the product of  $\varepsilon_{int.}(E) \times f_{geo}$  where  $\varepsilon_{int.} = f(E) < 1$ 

 $f_{geo}$ : geometrical factor between source-collimator-absorber  $\varepsilon_{int}$ : intrinsic detection efficiency

 $\epsilon_{FEP}$  and  $\epsilon_{int.}$  are difficult to calibrate accurately

### SMX3: A dedicated MMC for L X-ray spectrometry of actinides





- 4 absorbers of 1 mm<sup>2</sup>
   50 μm of Au + 17 μm of Ag thick
- Intrinsic efficiency > 99% between 10-25 keV

 $\leftarrow w$ 

- 10 − 20 s<sup>-1</sup> (τ<sub>d</sub> ≈ 4 ms)
- Energy resolution FWHM of 22 40 eV

sensor

# **Intrinsic Detection efficiency of AuAg absorbers**



ightarrow Quasi-constant intrinsic efficiency below 25 keV,  $arepsilon_{int.} \sim 1$ 

 $\rightarrow$  Minimize the efficiency correction

# Full energy peak detection efficiency calibration using <sup>241</sup>Am and MC simulations

$$\boldsymbol{\varepsilon}(\boldsymbol{E}) = \frac{N(E)}{\Delta t \cdot A \cdot I(E)}$$

# **Efficiency calibration**

- <sup>241</sup>Am spectrum to establish experimental data of
- Extendable dead applied to MMCs to determine  $\Delta t$
- Definition of a meta-geometry by and for Monte Carlo simulations

# Efficiency uncertainty

- < 10 keV</p>
  - ~ 4% uncertainty at 2 keV given by Be window
- 10 keV < E < 25 keV</li>
   0.7% uncertainty given by I(XLβ)
- > 25 keV
  - 1.2% to 2.4% uncertainty given by I(59.5 keV)



# L X-ray emission intensities of actinides

Many actinides decay by  $\alpha$ -emissions and have intense L X-ray PEIs between 10-25 keV



Nuclide	LX-ray intensity / 100 dis.	relative unc. (%)	Method	Number of measurements	calc. vs. exp.
Pu-239 → U-235+α	4.66	1.1	meas.	4	disagree
Pu-240 → U-236+α	10.34	1.5	meas.	6	agree
Am-241 → Np-237+α	37.66	0.5	meas.	9	disagree
Cm-242 → Pu-238+α	9.92	2.3	calc.	2	disagree
Cm-244 → Pu-240+α	8.92	2.6	calc.	1	agree
U-235 → Th-232+α	40.0	55.0	calc.	-	-





Cez

# **Data and spectrum processing**



Data continuously recorded with a 16 bits resolution DAQ
 @ 250-500 kHz

#### • Data analyzed offline

ullet

- Pulse triggering
- Live time determination
- Pulse energy estimation
- Pile-up rejection
- Temperature drift correction
- Non-linearity correction
- Energy resolution equalization
- Spectrum co-adding
- Counting statistics of few 10<sup>6</sup> X-ray counts
  - FWHM at best of 22 eV and 32 eV between 0 and 166 keV)

#### Spectrum processing

Diagram (solid lines) and satellites (dashed lines)

 R. Mariam *et al.* Spectrochim. Acta B Vol. 187, 2022, 106331
 19

 https://doi.org/10.1016/j.sab.2021.106331
 19



### **Comparison with PEIs in the literature**

#### Total L X-ray emission intensity



#### Siegbahn group L X-ray emission intensities

	$^{238}$ Pu( $\alpha$ )	$\rightarrow ^{234}\mathrm{U}$	$^{244}\mathrm{Cm}(\alpha)$	$\rightarrow ^{240}\mathrm{Pu}$	$^{233}$ Pa( $\beta^-$ ) $\rightarrow ^{233}$ U			
Group	This work	Johnston [34]	This work	Johnston [34]	This work	Calculated		
Lı	0.2418 (29)	0.231 (3)	0.2306 (35)	0.214 (3)	1.075 (19)	1.05 (4)		
Lα	3.816 (43)	3.81 (3)	3.49 (5)	3.38 (3)	15.69 (20)	16.9 (6)		
$L\eta$	0.1284 (16)	0.126 (2)	0.1002 (22)	0.102 (2)	0.235 (19)	0.272 (16)		
Lβ	5.23 (6)	5.18 (4)	4.22 (6)	4.08 (3)	16.89 (25)	18.1 (6)		
Lγ	1.291 (14)	1.29 (1)	1.023 (15)	0.991 (8)	3.97 (6)	4.23 (14)		

Agreement k = 1Agreement k = 2In disagreement

# **Tens of PEIs of individual L X-ray transitions**

Li	ne		<sup>238</sup> Pu	$(\alpha) \rightarrow {}^{234}\mathrm{U}$			$^{244}\mathrm{Cm}(\alpha) \rightarrow ^{240}\mathrm{Pu}$							
Siegbahn IUPAC		C Energy	X-ray emission intensity		Rel	<b>Rel. Unc. (%)</b>		Energy	X-ray emission intensity		Rel. Unc. (		(%)	
		(eV)	per 100 L X-rays	per 100 decays	$u_1$	$u_2$	<i>u</i> <sub>3</sub>	(eV)	per 100 L X-rays	per 100 decays	$u_1$	<i>u</i> <sub>2</sub>	<i>u</i> <sub>3</sub>	
-	L <sub>1</sub> -L <sub>3</sub>	4589.2	0.0214 (44)	0.00229 (47)	0.72	20	1.2	5054.63	0.0267 (13)	0.00242 (13)	0.37	5.0	1.6	
Lı	L <sub>3</sub> -M <sub>1</sub>	11618.4	2.257 (11)	0.2418 (30)	0.72	0.49	0.86	12124.4	2.540 (13)	0.2306 (36)	0.37	0.49	1.4	
Ls	L <sub>3</sub> -M <sub>2</sub>	11982.0	0.0227 (15)	0.00243 (16)	0.72	6.6	0.86	12503	0.0269 (20)	0.00244 (18)	0.37	7.4	1.4	
Lt	L <sub>3</sub> -M <sub>3</sub>	12864.7	0.0286 (17)	0.00307 (18)	0.72	5.8	0.86	13485.4	0.0315 (16)	0.00286 (15)	0.37	5.2	1.4	
La <sub>2</sub> La	L <sub>3</sub> -M <sub>4</sub>	13439.8	<b>3.604</b> (7) <b>32.007</b> (20)	<b>0.3862</b> (44) <b>3</b> <i>4</i> 29 (39)	0.72	0.20	0.86	14074.5	<b>3.806</b> (11) <b>34</b> 68 (7)	<b>0.346</b> (5) <b>3</b> 149 (47)	0.37	0.29	1.4	
$La_1$	L <sub>3</sub> -1V15	13014.0	52.007 (20)	3.429 (39)	0.72	0.002	0.00	(2022) 10622	34.00 (7)	5.149 (47)	0.57	0.21	1.4	
$L\eta$	$\mathbf{L}_{\mathbf{i}}$		Зр	ectrochinica Acta Part B.	Atomic a	pectros	opy 187	(2022) 10033	1		0.37	1.6	1.4	
	-	ANA DAKA LUL-		Contents lists	availab	a at Sa	ioncoDir	last			0.07	1.0		
Lβ <sub>6</sub> ι α				Contents lists	availab	e at so	lenceDir	ect		SPECTROCHIMICA	0.37	1.2	1.4	
$L \rho_{2,15}$	L3	a Alexandre	0	1.1	Dent	D. A				ACTA server reason	0.57	0.23	1.4	
L <i>B</i> ,	L		Spectroc	nimica Acta	Part	<b>В:</b> А	tom	ic Spect	roscopy		0.37	1.9	1.4	
$L\beta_{17}$	L	A les									0.37	2.5	1.4	
Lu	L <sub>3</sub> FI S	FVIER		iournal homepage	www.e	elsevie	r.com/lc	ocate/sab			0.27	25	14	
$L\beta_7$				,							0.57	5.5	1.4	
L $meta_{s}$	L <sub>3</sub>										0.37	0.81	1.4	
- 0	L											0.00		
$L\beta_1$		•						c 238-	244	Check for	0.37	0.10	1.4	
$Lp_3$	L Det	erminat	ion of L-X ray a	absolute emiss	sion	nter	isitie	s of 2001	Pu, ~' 'Cm,	updates	0.37	1.4	1.4	
$L\beta_{10}$	L 237	Nn and	233 Da radionucl	ides using a n	notal	lic m	naan	atic calc	rimeter		0.37	14	1.4	
$Lp_9$		np and	ra laulolluci	lues using a n	iictai	ne n	lagin	enc car	milleter		0.57	0.5	1.4	
Lи	L		abaa	a *				2 1/ .	- 9		0.37	11	14	
-	L Riha	am Marian	n <sup>a, 9</sup> , Matias Rodrig	ies", Martin Loi	dl <sup>a</sup> , S	ylvie	Pierre	e", Valérie	Lourenço "		0.37	4.4	1.4	
Lĸ	L <sup>a</sup> Unive	rsité Paris-Saclay	CFA List Laboratoire National	Henri Recauerel (LNF-LNHR)	F-91120	Palaiseau	France				0.27	0.27	0.3	
L <sub>2</sub>	L <sup>b</sup> Unive	rsité Paris-Saclay	CNRS/IN2P3, IJCLab, 91405 O	rsay, France	. ,	unanocun,	Trance				0.37	4.6	1.4	
$L\gamma_8'$	$L_2-N_6$	20556.1	0.0010(8)	0.00660 (11)	0.72	1.3	0.86	21829.3	0.0607 (21)	0.00551 (21)	0.37	3.4	1.4	
$L\gamma_8$	$L_2 - O_1$	20625.7	0.0910 (11)	0.00975 (16)	0.72	1.2	0.86	21914	0.0646 (18)	0.00586 (18)	0.37	2.8	1.4	
Lγ <sub>3</sub>	$L_1 - N_3$	20712.4	0.0594 (13)	0.00636 (15)	0.72	2.1	0.86	21980	0.0665 (18)	0.00604 (18)	0.37	2.6	1.4	
$L\gamma_6$	$L_2-O_4$	20842.0	1.948 (6)	0.2087 (18)	0.72	0.33	0.36	22149.1	1.911 (9)	0.1735 (27)	0.37	0.45	1.4	
-	$L_2$ - $P_1$	20904.0	0.0117 (17)	0.00126 (18)	0.72	15	0.86	-	-	-	-	-	-	
-	$L_2-P_{4,5}$	20941.7	0.03312 (50)	0.00355 (7)	0.72	1.5	0.86	22260.9	0.0343 (11)	0.00312 (11)	0.37	3.2	1.4	
$L\gamma_4'$	$L_1-O_2$	21498.1	0.01838 (41)	0.001969 (49)	0.72	2.2	0.85	22823.3	0.0141 (7)	0.00128 (7)	0.37	5.0	1.4	
Τ	τo	01564.0	0.01245(24)	0.001441 (20)	0.70	0.5	0.05	22000.0	0.01(0)	0.00147 (7)	0.27	1 (	1 4	

21

# **Limits of previous measurements**

- Previous PEI measurements limited by the uncertainty on the FEP detection efficiency
- Itself limited by the recommended values of the PEIs of <sup>241</sup>Am used for the efficiency calibration



PEIs are ultimately interdependent and correlated to some extent

# **Absolute L X-ray PEIs of <sup>241</sup>Am independently of other PEIs**





#### Conditions:

```
    ✓ f_{geo,\alpha} = f_{geo,ph.}
    ✓ 100% decay by α-particle emission
    ✓ ε_{int,\alpha} ≈ ε_{int,ph.} ≈ 1
```



 $\varepsilon_{int}$ : Intrinsic efficiency  $f_{geo,ph}$ : geometrical efficiency

However measuring  $\alpha$ -particles and photons in the same spectrum is not possible:

- × MMC sensitivity for  $\alpha$ -particles (few MeV) and X-rays (10s of keV) must be different
- × Ten times as many emitted particles per decay as there are X-rays
  - $\rightarrow$  Many pile-up or reduced counting statistics, electrons/X-ray interferences

# **Measurements of two <sup>241</sup>Am sources with different activities**

#### 1<sup>st</sup> measurement: $\alpha$ spectrum

- <sup>241</sup>Am source of 1.8 kBq
- Lower MMC sensitivity
- No Be window
- FWHM resolution of 3.3 keV





# **Measurements of two <sup>241</sup>Am sources with** different activities

#### 1<sup>st</sup> measurement: $\alpha$ spectrum

- <sup>241</sup>Am source of 1.8 kBq
- Lower MMC sensitivity
- No Be window
- FWHM resolution of 3.3 keV

#### 2<sup>nd</sup> measurement: X-ray spectrum

- <sup>241</sup>Am source of 32 kBq
- High MMC sensitivity
- Be window to stop the  $\alpha$ -particles
- spectrum FWHM resolution of 28 eV





 $\rightarrow n_{\alpha}$ 

#### L X-ray spectrum of <sup>241</sup>Am( $\alpha$ ) $\rightarrow$ <sup>237</sup>Np Total fit 100000 L₁ diagram L<sub>2</sub> diagram 10000 L<sub>2</sub> satellite Other 1000 100 19500 20000 20500 21000 21500 22000 22500 100000 10000 е c Esc. L Ag loo0 Esc. L A Counts 11500 12000 12500 13000 14500 13500 14000 15000



# Additional corrections due to the measurement of 2 sources

$$I(E) = \frac{n_{FEP}(E)}{n_{\alpha}} \frac{F_A \cdot F_{source} \cdot \varepsilon_{int,\alpha}}{\varepsilon_{int,ph}(E) \times t_{Be}(E)}$$

- $n_{FEP}$  and  $n_{\alpha}$  from energy spectra
- $F_A$  ratio between source activities determined by conventional  $\alpha$ -particles spectrometry
- *F*<sub>source</sub> correction factor for the inhomogeneity of the surface source activity determined by radioactive source imager.



- $\varepsilon_{int,\alpha}$  and  $\varepsilon_{int,ph}$  intrinsic efficiencies ~ 1, determined by Monte Carlo simulations.
- $t_{Be}$  transmission through Be window, calculated.

# Results of L X-ray PEIs from <sup>241</sup>Am( $\alpha$ ) $\rightarrow$ <sup>237</sup>Np

## Total L X-ray and Siegbahn group (L $\alpha$ , L $\beta$ , L $\gamma$ ...) PEIs

- Good agreement with all the previous published data
- Relative uncertainty (0.32%) 2 times lower than the most precise measurement



# **Results of L X-ray PEIs from {}^{241}Am(\alpha) \rightarrow {}^{237}Np**

#### 33 PEIs of individual X-ray transitions are provided

				Relative uncertainties					Relative uncertainties			
X-ray tra	insition	Energy	PEI per 100				Siegbahn	PEI per				
UIPAC	Siegbahn	(eV)	decays	Total	Fitting procedure	Counting statistics	group	100 decays	Total	Fitting procedure	Counting statistics	
L <sub>1</sub> -L <sub>3</sub>	-	4820	0.2289 (15)	0.65%	0.41%	0.42%						
$L_3-M_1$	Lı	11873	0.8989 (38)	0.42%	0.13%	0.21%						
$L_3-M_2$	Lt	12250	0.01024 (26)	2.57%	0.64%	1.96%						
L <sub>3</sub> -M <sub>3</sub>	Ls	13179	0.0101 (8)	7.59%	7.33%	1.98%						
L <sub>3</sub> -M <sub>4</sub>	$L\alpha_1$	13762	1.2581 (49)	0.39%	0.19%	0.18%	Ια	13 046 (41)	0.31%	0.084%	0.060%	
L_3-M_5	$L\alpha_2$	13944	11.788 (38)	0.32%	0.13%	0.06%	La	13.040 (41)	0.31%	0.084%	0.009%	
$L_2-M_1$	$L\eta$											
$L_3-N_1$	-	IOP Publishing 1 F	Bureau International des Poio	ts et Mesures					Metrolo	ngia		
$L_{3}-N_{4,5}$	$L\beta_{2,15}$	Metrologia 60 (2022) 0	025005 (19pp)					https://doi.org/10.1099	/1691-7575/ach			
$L_1-M_2$	$L\beta_4$	Metrologia 00 (2023) 0	23003 (Topp)					maps.//doi.org/10.1066	/1001-7575/800	551		
$L_{3}-N_{6,7}$	$L\beta_7'$											
L <sub>3</sub> -O <sub>1,2,3</sub>	$L\beta_7$	Dotorn	nination		hool	Ito N	n I v	KOV				
L <sub>3</sub> -O <sub>4,5</sub> - L <sub>3</sub> -	LBe	Delen	IIIIalioi		102011	le n		ray		64%	0.058%	
$P_{1,4,5}$	1-3			! . !	<b>f</b>	24	1	-				
$L_2-M_4$	$L\beta_1$	emiss	ion inte	nsiti	es tro	om ~ '	'AM	decay				
$L_2-M_5$	-	_										
$L_1 - M_3$	$L\beta_3$	usina	a metal	lic m	adne	tic ca	alorin	neter				
$L_1 - M_4$	$L\beta_{10}$	domg	u motu									
$ L_1 - M_5$	$\frac{L\beta_9}{L}$											
$L_2 - N_1$	$L\gamma_5$	Matias Rodrig	ques*©, Martin L	oidl and S	Sylvie Pierre	0						
$L_2 - N_3$	- I.v											
$L_2 - N_4$	$L\gamma_1$	Université Paris-Sa	clay, CEA, List, Laborat	toire National	Henri Becquerel	(LNE-LNHB)	, F-91120					
$L_1 - N_2$	$L_{\gamma_2}$	Palaiseau, France										
$L_2 = \Gamma_6$		21260	0 02841 (48)	1 69%	1 17%	1 18%						
	L78 L%	21200	0.02041 (40)	0.46%	0.20%	0.30%						
L <sub>1</sub> -O.	Lγ.	21489	0.6260 (30)	0.48%	0.28%	0.25%	Lγ	4.883 (20)	0.41%	0.26%	0.11%	
L <sub>2</sub> 0 <sub>4</sub>	-76	21555	0.00386 (16)	4.02%	2.41%	3.20%						
	-	21595	0.02246 (31)	1.39%	0.28%	1.33%						
$L_1 - N_5$	-	21656	0.02041 (30)	1.46%	0.32%	1.39%						
$L_1 - O_2$	L <sub>V4</sub> '	22155	0.1139 (8)	0.67%	0.15%	0.59%						
$L_1 - O_3$	$L\gamma_{4}$	22216	0.1057 (7)	0.70%	0.15%	0.61%						
L <sub>1</sub> -O <sub>4</sub> =	/ <del>+</del>	22319	0.00639 (16)	2.58%	0.61%	2.49%						
$L_1 - P_{2,3}$		22404	0.04361 (46)	1.05%	0.34%	0.95%						

28

cea

# **Conclusions and perspectives**



#### MMCs are useful tool to provide accurate decay data in metrology of ionizing radiation

- MMCs are suitable for decay data measurements by DES (< 200 keV)
  - Beta spectrum shape
  - End point energies
  - Beta branch probabilities
  - EC probabilities

 $\rightarrow$  We are currently developing multiple MMC channels to achieve high statistics (108 counts) for beta spectrum of <sup>129</sup>I

- MMCs are suitable for precise and detailed absolute PEI determinations (<100 keV)
  - L X-ray PEIs for <sup>238</sup>Pu, <sup>244</sup>Cm, <sup>233</sup>Pa and <sup>237</sup>Np with relative uncertainties of ~ 0,8%
  - L X-ray PEIs for <sup>241</sup>Am without efficiency calibration based on other PEIs with relative uncertainties of ~0.3%
  - $\gamma$ -ray and X-ray PEIs in the range 25 keV-100 keV with relative uncertainties of ~ 0,8%-1%
  - → We are currently developing an MMC array for absolute PEIs of photons < 10 keV
- Measuring these decay data at higher energies is challenging:
  - due to loss of Bremsstrahlung photons for electrons
  - due to loss of efficiency for photons





# Thank you for your attention

Matias Rodrigues

