

LOW
RADIOACTIVITY
TECHNIQUES



2024

WORKSHOP IX



Developments, features and perspectives of crystal scintillators of the Cs_2MCl_6 family (M = Hf or Zr) to search for rare processes

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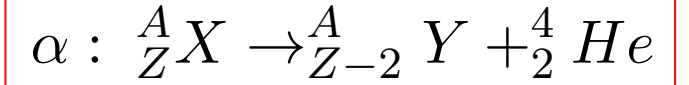
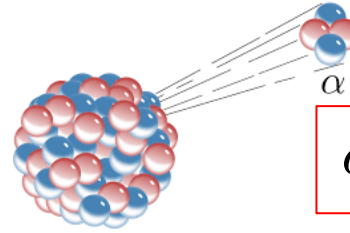
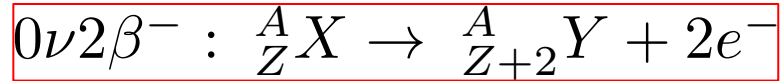
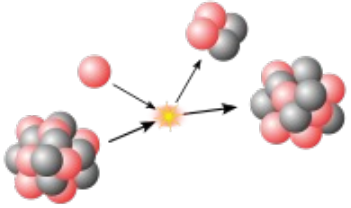
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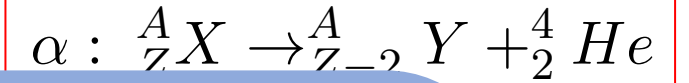
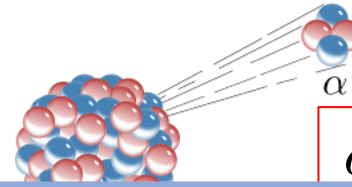
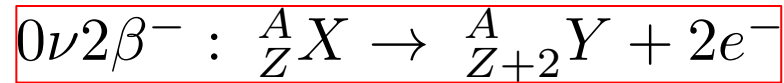
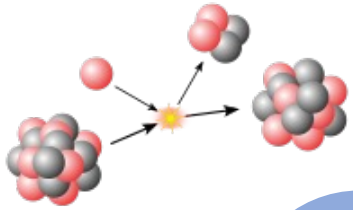
Interest in studying the 2β and rare α decay



- ❖ $2\nu 2\beta$ decay allowed in the SM → lepton number L conserved
- ❖ $0\nu 2\beta$, if observed, could open a new window beyond the SM → L violated ($\Delta L = 2$) → massive Majorana neutrino.
- ❖ Current sensitivity for $2\nu 2\beta^-$ decay: $T_{1/2} \sim 10^{18} - 10^{24}$ yr; for $0\nu 2\beta^-$: $T_{1/2} \sim 10^{24} - 10^{26}$ yr.
- ❖ To test, e.g., calculations of different nuclear shapes and the decay modes that involve the vector and axial-vector g_A weak effective coupling constants.

- ❖ Details on the nuclear structure, the nuclear levels and the properties of nuclei.
- ❖ Rare α decay: $T_{1/2} > 10^{14}$ yr.
- ❖ Essential also for nuclear and particle astrophysics studies (α -capture reactions, β -delayed fission, nucleosynthesis).

Interest in studying the 2β and rare α decay

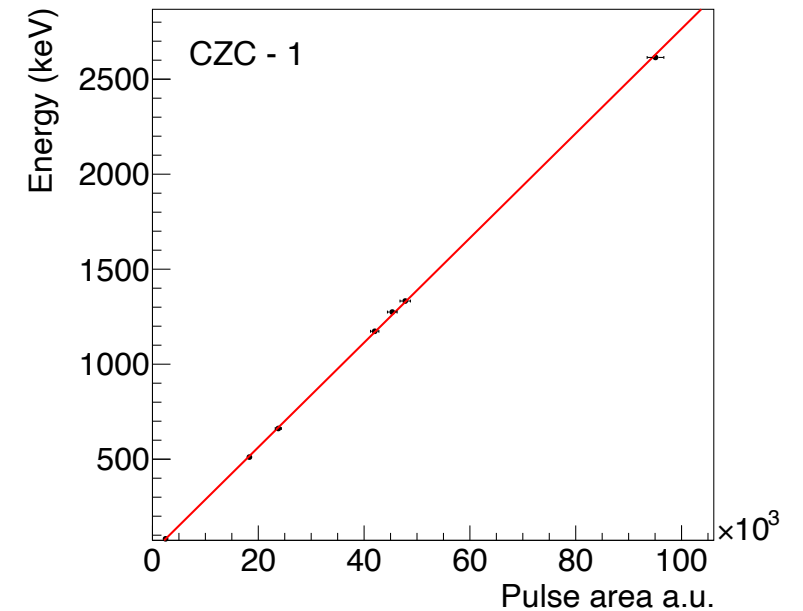


Requirements: high radiopurity (e.g. $\lesssim 0.1$ ppb), high-sensitivity (energy threshold \sim keV), good detector performances (efficiency $> 90\%$), well known technology, stability over the running conditions (variation $< 1\%$), acceptable cost (1,000 - 10,000 Eur/kg), safety, many isotopes and decay modes explorable etc.

- ❖ $2\nu 2\beta$ decays conserve lepton number, the structure, the of nuclei.
- ❖ $0\nu 2\beta$, if SM \rightarrow $L\nu$
- ❖ Current particle reactions, for $0\nu 2\beta$ (s).
- ❖ To test, e.g., calculations of different nuclear shapes and the decay modes that involve the vector and axial-vector g_A weak effective coupling constants.

Some general properties	Cs ₂ HfCl ₆	Cs ₂ ZrCl ₆
Effective atomic number	58	46.6
Density (g/cm ³)	3.9	3.4
Melting point (°C)	820	850
Crystal structure	Cubic	Cubic
Emission maximum (nm)	400 - 430	450 - 470
Scintillation time constants (μs)	0.4; 5.1; 15.2 *	0.4; 2.7; 12.5*
Light Yield	up to 30000 photons/MeV**	up to 41000 photons/MeV**
Linearity of the energy response	Excellent, down to 100 keV	Excellent, down to 100 keV
Energy resolution (FWHM, %) @ 662 keV	3.2 - 3.7***	3.5 - 7.0***
Pulse-shape discrimination ability	Excellent	Excellent
Mass fraction of isotope of interest (%)	27	16

Cs₂HfCl₆ (CHC) and Cs₂ZrCl₆ (CZC) crystal scintillators



* for alpha events at room temperature (*Dalton Trans. 2022, 51, 6944-6954*)

** for gamma quanta at room temperature

*** depends on the crystal quality, surface treatment and readout system

Production and growth of Cs_2HfCl_6 crystals

Produced at **Queen's University, Canada**

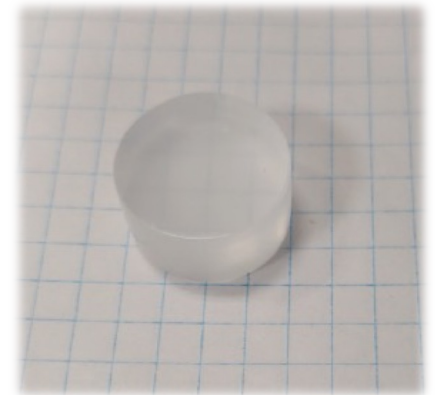
CHC (6.90 g)

- ✓ CsCl (99.998%) + HfCl_4 (99.8%) as starting materials.
- ✓ HfCl_4 powder subjected to a three-fold purification process.
- ✓ Grown by **Bridgman technique**: growth at 5° C/cm, at 1 cm/day).

CHC (16.87 g)

- ✓ CsCl (99.9%) + HfCl_4 (98%) as starting materials.
- ✓ HfCl_4 powder subjected to a three-stage sublimation process.
- ✓ Grown by **vertical Bridgman technique**: «fast» growth (25° C/cm, at 1.46 mm/hours) + «slow» growth (20° C/cm, at 0.5 mm/hour).

Ø21.20(5)×12.8(1) mm



Then polished with 1200 grit sandpaper, mineral oil as lubricant, cleaned by toluene.

Cs₂HfCl₆ chemical contaminants

measured by ICP-MS analysis.

The limits are
at 68% C.L.

Nuclide	Concentration (ppb)
	CHC
	6.90 g [1]
¹⁴⁴ Nd	<2.4
¹⁴⁷ Sm	0.6(1)
¹⁴⁸ Sm	0.4(1)
¹⁵¹ Eu	19(7)
¹⁵² Gd	<0.02
¹⁸⁰ W	<0.4
¹⁸⁴ Os	<0.003
¹⁸⁶ Os	<0.25
¹⁹⁰ Pt	<0.02
²⁰⁹ Bi	<2

Element	Concentration (ppb)
	CHC
	16.87 g [2]
U	0.73(22)
Th	0.16(5)
Pb	440(130)
Sm	2(1)
K	1900(570)

[1] V. Caracciolo et al.,
NPA 1002 (2020) 121941.

[2] P. Belli et al. 2024, to
appear

Cs₂HfCl₆ crystal radiopurity

measured with the ultra-low background **HP-Ge** γ spectrometers of the **STELLA** facility at LNGS over ~ 700 hours.

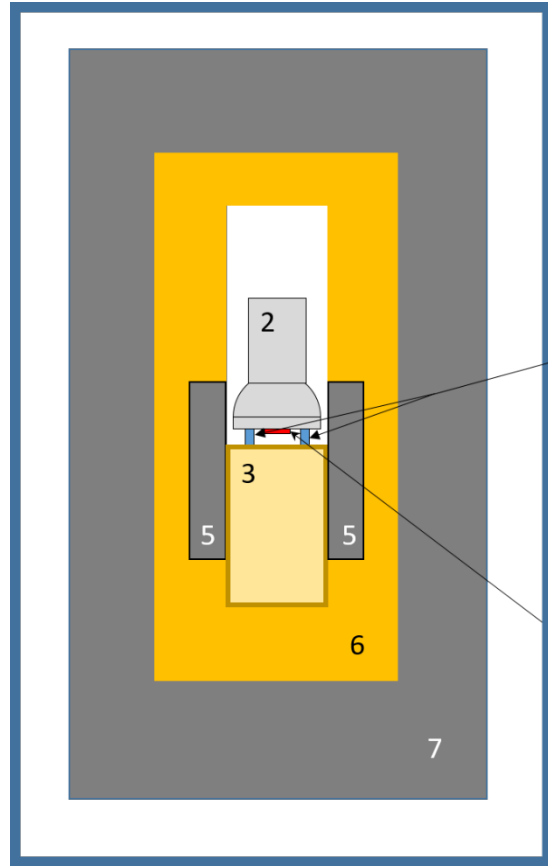
Chain	Nuclide	Activity (mBq/kg)		
		CHC		
		6.90 g [1]	16.87 g [2]	
²³⁸ U	²²⁶ Ra	<23	<13	} Natural
	²³⁴ Th	<0.80	<1200	
	^{234m} Pa	<0.48	<18	
²³⁵ U	²³⁵ U	<14	<18	
²³² Th	²²⁸ Ra	<12	<13	
	²²⁸ Th	<3.6	<17	
	²⁰² Pb	<9.1	-	
	¹⁹⁰ Pt	<20	-	
	¹⁸¹ Hf	<11	-	
	¹³⁷ Cs	$0.78(8) \times 10^3$	<10	→ Artificial
	¹³⁴ Cs	79(8)	37(4)	} Cosmogenic activation
	¹³² Cs	<15	-	
	⁶⁰ Co	<25	-	
	⁴⁴ Ti	10(4)	-	
	⁴⁰ K	$0.4(1) \times 10^3$	<240	→ Natural

Only land transportation!
 $T_{1/2} \approx 2$ years

[1] V. Caracciolo et al., NPA 1002 (2020) 121941.

[2] P. Belli et al. 2024, to appear

First low-background measurement of CHC at LNGS

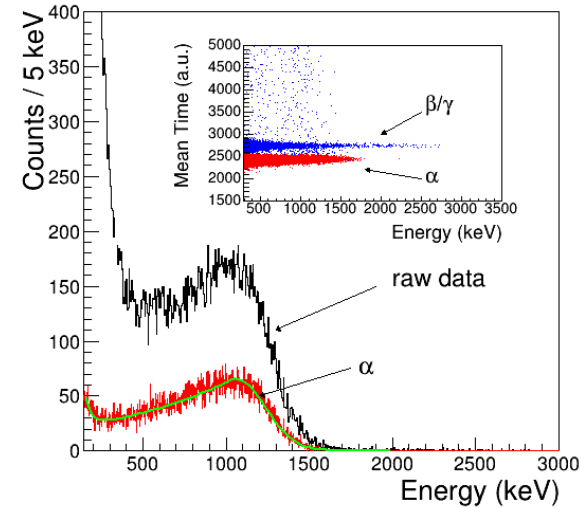


- ✓ CHC crystal (6.90(1) g) coupled low-radioactivity PMT (Hamamatsu R6233MOD) placed above the end-cap of the ultra-low background HP-Ge
- ✓ CAEN DT5720B digitizer 250 MSamples/s;
- ✓ 2848 h data taking

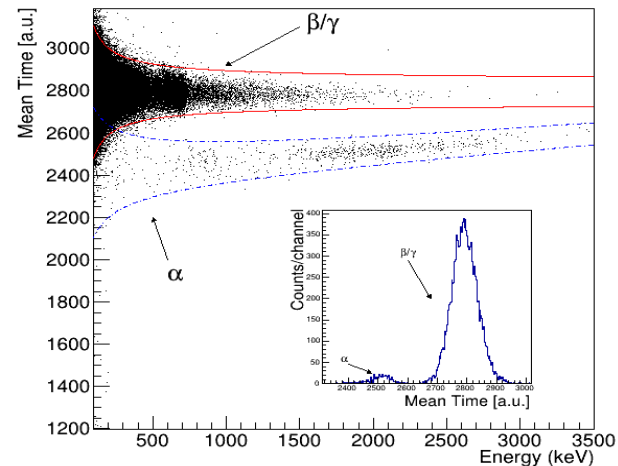
STELLA facility of the LNGS

- (1) CHC crystal scintillator
- (2) PMT
- (3) HP-Ge detector
- (4) Teflon ring
- (5) Pb, 2.5 cm
- (6) HP Cu
- (7) Pb, 25 cm
- (8) Plexiglas box

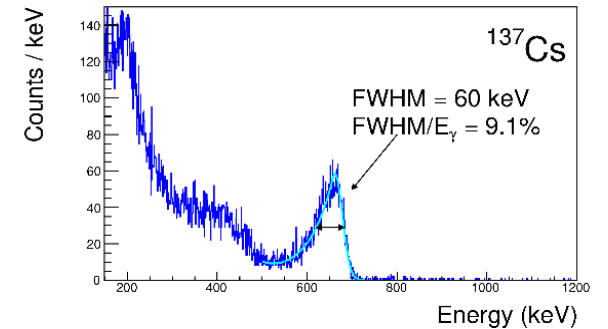
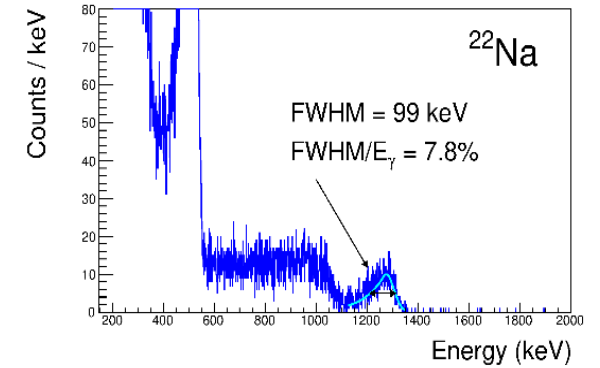
Preliminary quenching factor (QF) estimation → ²⁴¹Am



Selection efficiency is 99% in [0.4–3.0] MeV



Energy resolution = 9.1% @ 662 keV, γ

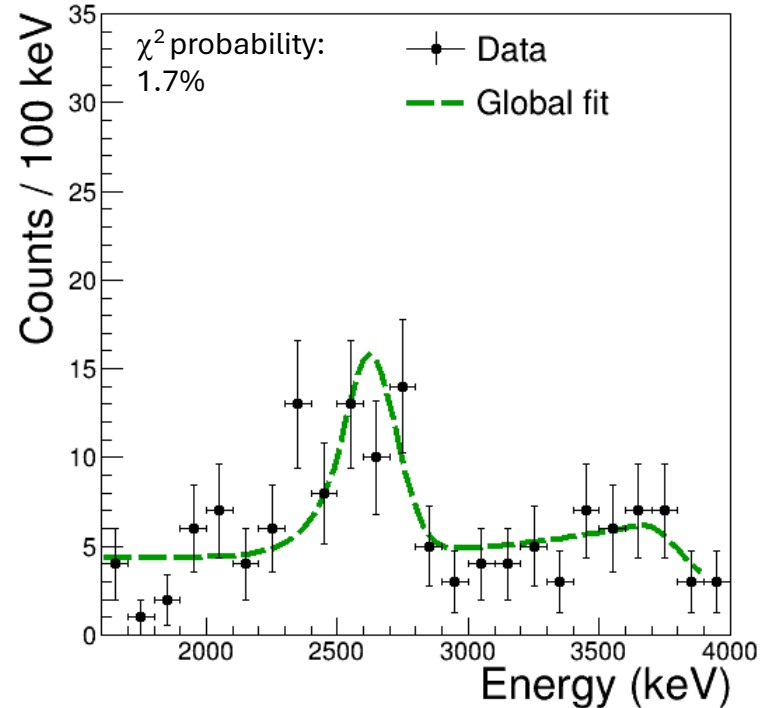
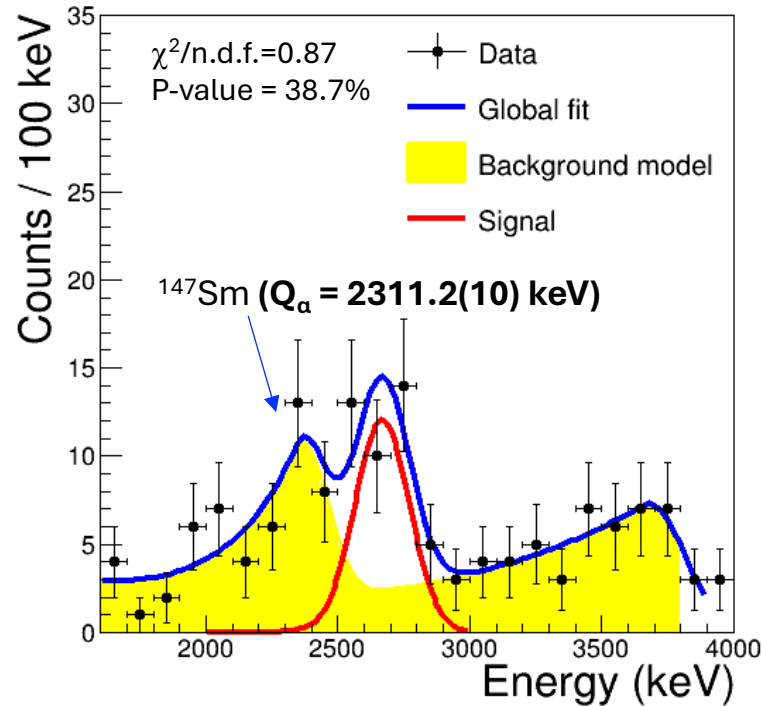


Fit of the α spectrum of CHC crystal and results

Energy spectrum of the α events selected by PSD over 2848 h.

Signal: α decay of ^{174}Hf ($Q_\alpha = 2494.5(2.3)$ keV)

The total internal α activity is **7.8(3) mBq/kg**.



In **2848** h of data taking, the **measured 553(23) α events** (expected number = **1100 counts**) **rule out the old result of $2.0(4) \times 10^{15}$ yr** (*T.P. Kohman, Phys. Rev. 121, 1758, 1961*) for $T_{1/2}$ of a decay of ^{174}Hf .

$T_{1/2} = (7.0 \pm 1.2) \times 10^{16}$ yr

New measurement of the α decay of ^{174}Hf to the g.s. of ^{174}Yb

Fit with 2 gaussians + pol1

Parameters of the fit:

First peak

Area = (36 ± 15) counts

Peak position = (787 ± 18) keV

Activity of ^{147}Sm = (0.25 ± 0.10) mBq/kg,
corresponding to a concentration of (2.0 ± 0.8) ppb
of $^{\text{nat}}\text{Sm}$, in agreement with ICP-MS measurements.

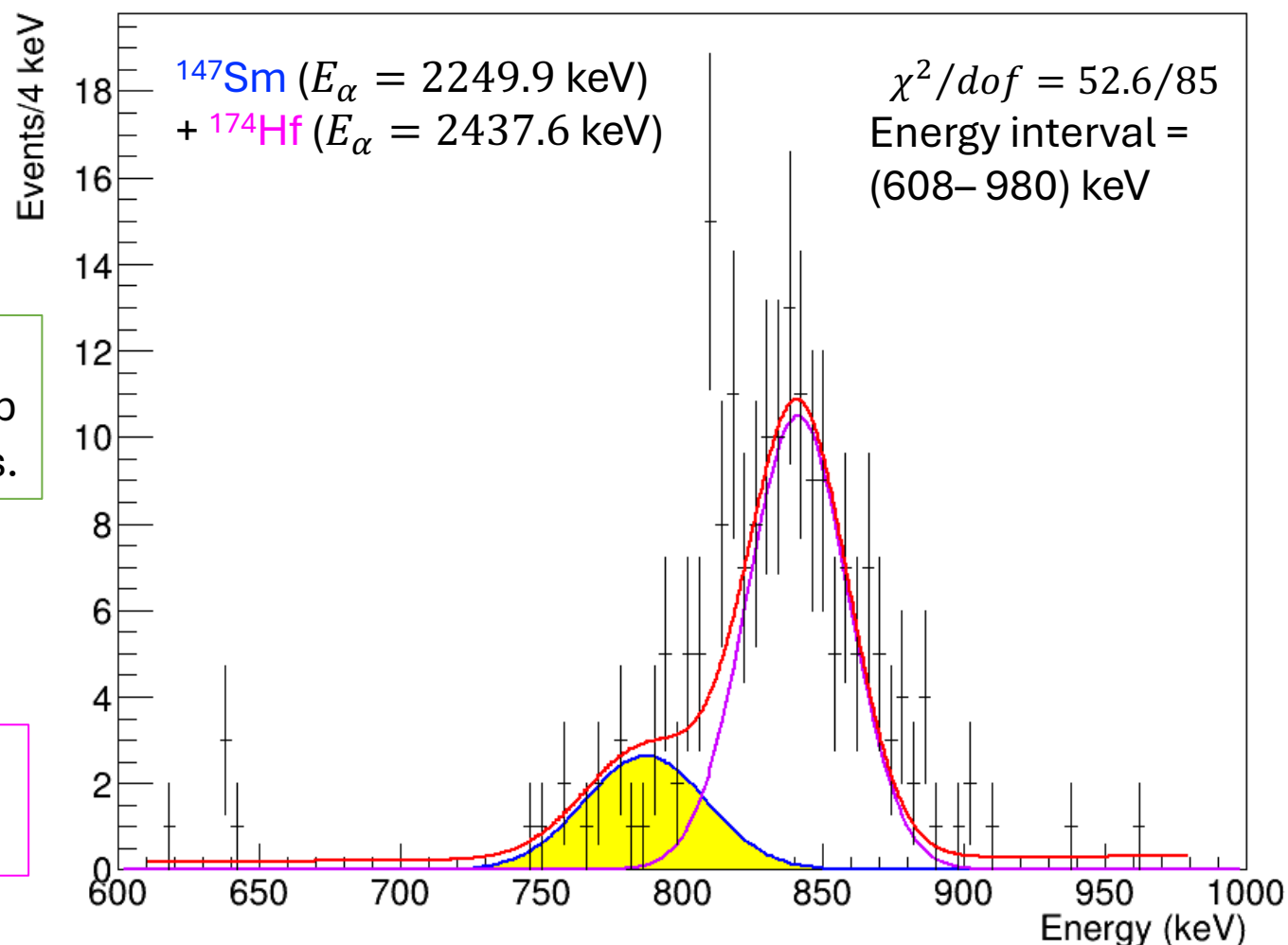
Second peak

Area = (118 ± 37) counts

Peak position = (841 ± 3) keV

$T_{1/2} = 3.8_{-0.9}^{+1.7} \times 10^{16}$ yr of α decay of ^{174}Hf
to the g.s. of ^{174}Yb

➤ Currently, 4 CHC crystals ($\varnothing 26 \times 20$ mm³) encapsulated in silicone-based sealant, **are ongoing** to improve sensitivity on α decay of ^{174}Hf .

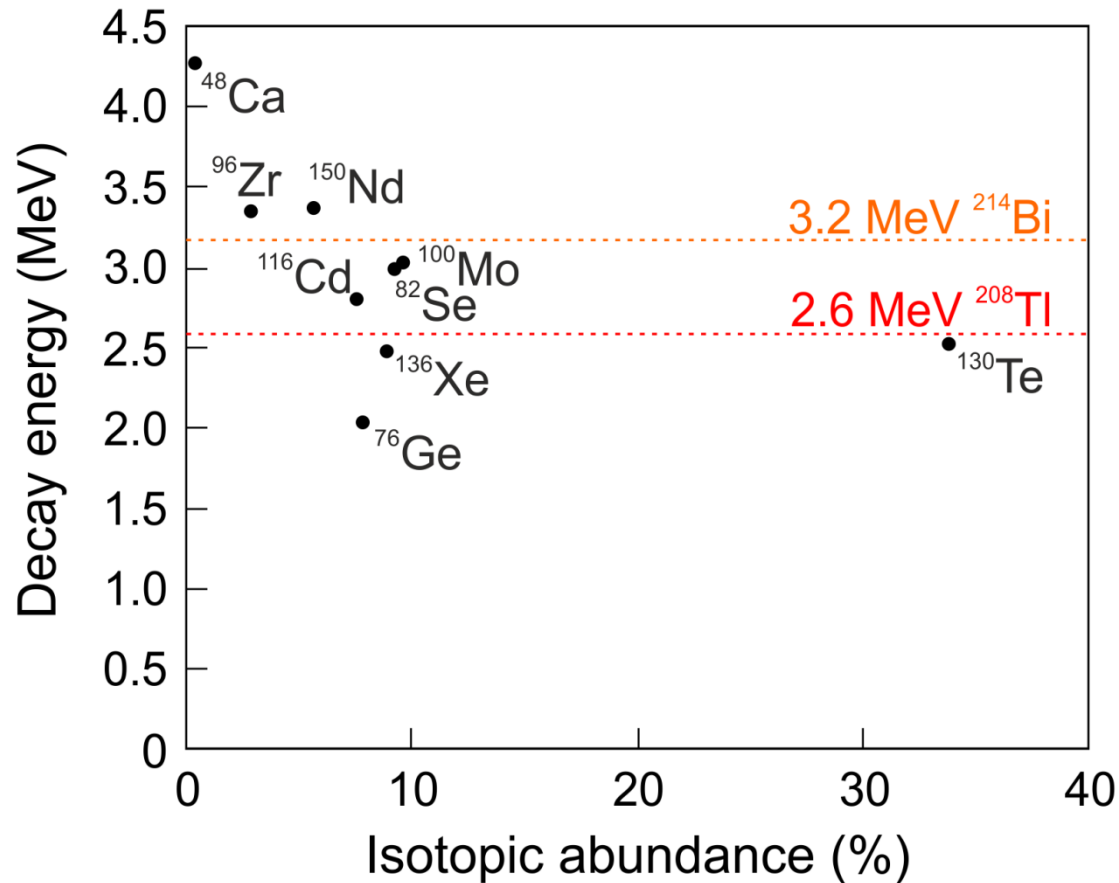


Q.F. = 0.350 ± 0.008 for ^{147}Sm

Q.F. = 0.345 ± 0.001 for ^{174}Hf

$0\nu 2\beta$ searches with non-trivial candidates

There are more than 60 potentially 2β -active isotopes, but only few of them are currently under consideration



^{76}Ge , ^{130}Te , ^{136}Xe are facing issues with an internal and environmental gamma background, while profiting from well-developed crystal production and material purification technologies.

^{82}Se , ^{100}Mo , ^{116}Cd – only ^{100}Mo is under consideration due to a well-developed detector material and its high radiopurity.

^{48}Ca , ^{96}Zr , ^{150}Nd are the less studied due to combination of unfavorable experimental conditions specific to each of them.

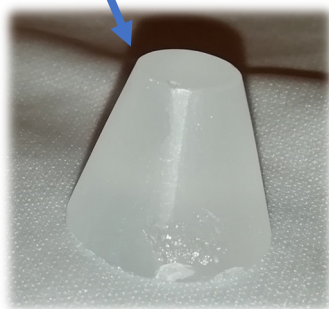
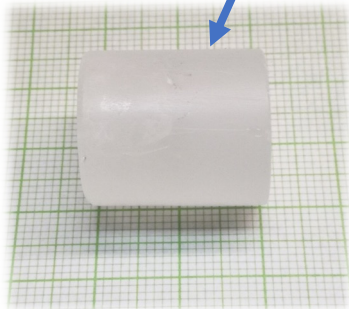
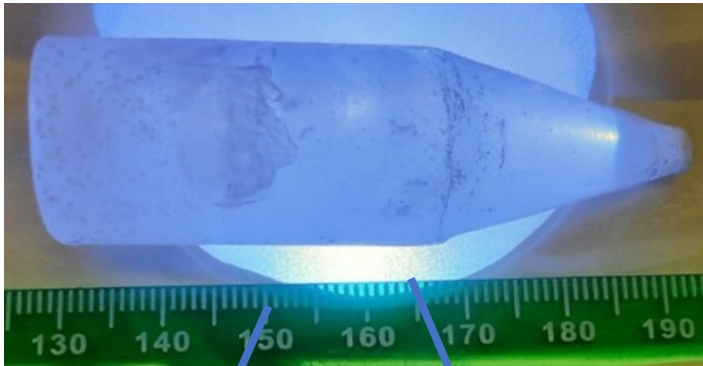
Recent proposal: to study $0\nu 2\beta$ of ^{96}Zr with novel Cs_2ZrCl_6 scintillators via “source = detector” experimental approach

- $Q_{\beta\beta} (^{96}\text{Zr}) = 3.35 \text{ MeV}$
- Favorable from a theoretical point of view $T_{1/2} \sim (Q_{\beta\beta})^5$
- Reasonable natural i.a. (2.8%)
- New advanced detector material (Cs_2ZrCl_6)
- Crystal production under full control
- Extensive studies of the detector properties

Production and growth of Cs_2ZrCl_6 crystals

Produced at **Queen's University, Canada**

Ø21.5×60 mm, about 60 g



CZC cone (10.63 g) & cylinder (23.95 g)

- ✓ CsCl (99.9%) + ZrCl_4 (99.9%) as starting materials
- ✓ ZrCl_4 powder subjected to a two-stage sublimation process
- ✓ Grown by **vertical Bridgman technique**: «fast» growth (28° C/cm, at 1.5 mm/hour) + «slow» growth (25° C/cm, at 0.5 mm/hour)

CZC -1 (19.21 g), CZC - 2 (19.86 g), CZC - 3 (20.43 g)

- ✓ CsCl (99.9%) + ZrCl_4 (99.9%) & CsCl (99.999%) + ZrCl_4 (99.99%) as starting materials
- ✓ ZrCl_4 powders subjected to a two-stage sublimation process
- ✓ Grown by **vertical Bridgman technique**: «fast» growth 20° C/cm, at 24 mm/day) + «slow» growth (25° C/cm, at 12 mm/day).
- ✓ Then the single crystalline boules cooled-down to room temperature with a temperature gradient of 0.1°C/min.

+ encapsulated
using SYLGARD
184™ Silicone
Elastomer Kit

Then polished with 1200 grit sandpaper, mineral oil as lubricant, cleaned by toluene.

Chemical purity of reagents at each production stage

HR-ICP-MS, concentrations are in ppb with 25% uncertainty

	CsCl initial	ZrCl ₄ initial	ZrCl ₄ 1st sublimation	ZrCl ₄ 2nd sublimation	CZC 1st growth, tail	CZC 1st growth, nose	CZC 2st growth, middle
K	300	15000	700	700	2500	200	500
La	0.7	1.5	1	1	1	0.6	0.6
Ce	1.5	2	1	1	2.5	3	2
Pr	0.1	4	6	6	1.5	1	1
Nd	<1	30	25	30	5	3	3
Sm	0.5	1	4	1	1	0.6	0.6
Eu-Lu	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5	<0.5
Hf	35	6400	5200	5600	1200	1800	1600
Ta, W, Re, Os, Ir	<2	<2	<2	<2	<2	<2	<2
Pt	<1	<100	<100	<100	<25	<25	<25
Tl	0.4	<0.2	<0.2	0.2	1	<0.2	<0.2
Pb	<1	30	20	30	150	1	13
Bi	<0.5	<0.5	1.5	2.6	1.5	<0.5	1.6
Th	<0.05	70	0.5	0.2	<0.05	<0.05	<0.05
U	<0.05	1000	7	0.36	0.35	0.13	<0.05

Cs₂ZrCl₆ crystal radiopurity

measured with the ultra-low background **HP-Ge** γ spectrometers of the **STELLA** facility at LNGS over 700 hours.

Chain	Nuclide	Activity (mBq/kg)	
		Cone	Cylinder
		10.63 g	23.95 g
²³⁸ U	²²⁶ Ra	60(10)	< 8.7
	²³⁴ Th	< 180	< 260
	^{234m} Pa	< 630	< 160
	²³⁵ U	< 16	< 12
²³² Th	²²⁸ Ra	< 16	< 23
	²²⁸ Th	< 6.7	< 8.2
	¹³⁷ Cs	< 7.1	< 1.6
	¹³⁴ Cs	49(6)	42(5)
	¹³² Cs	< 8.2	< 11
	⁴⁰ K	<120	<95

Surface cross-contamination happened during the sample preparation and installation.

Natural

Artificial

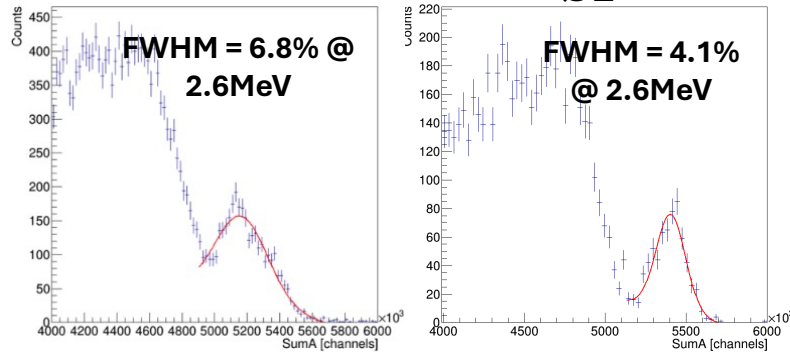
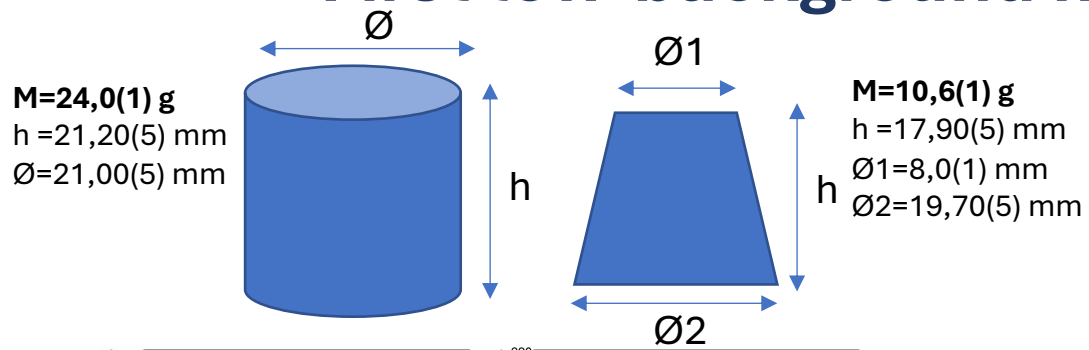
Cosmogenic activation

Natural

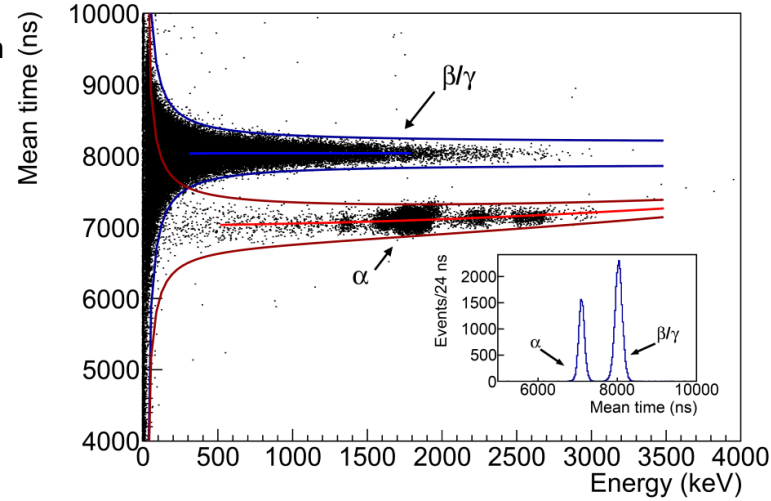
Only land transportation!
T_{1/2} ≈ 2 years

Our crystals are rather clean, even if they were grown from 99.9% purity grade raw materials

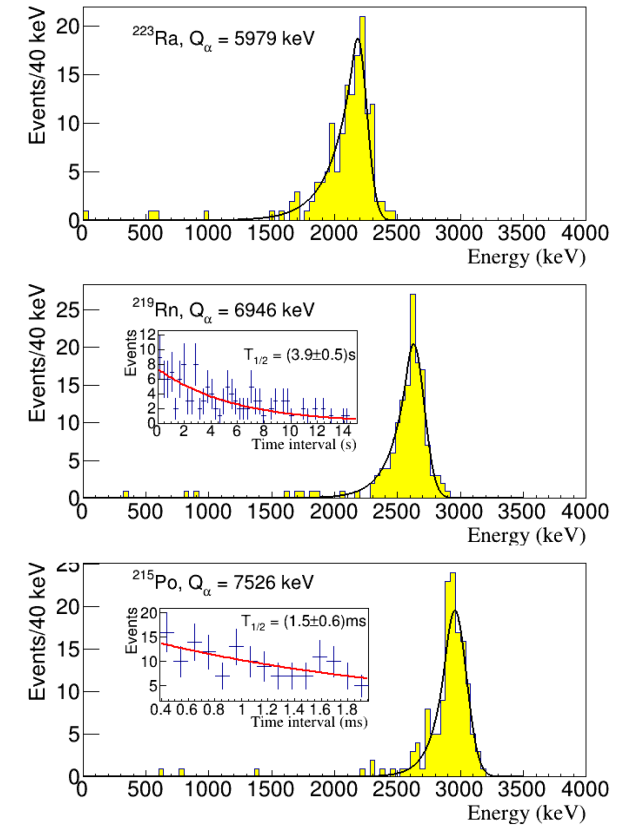
First low-background measurements of CZC at LNGS (Italy)



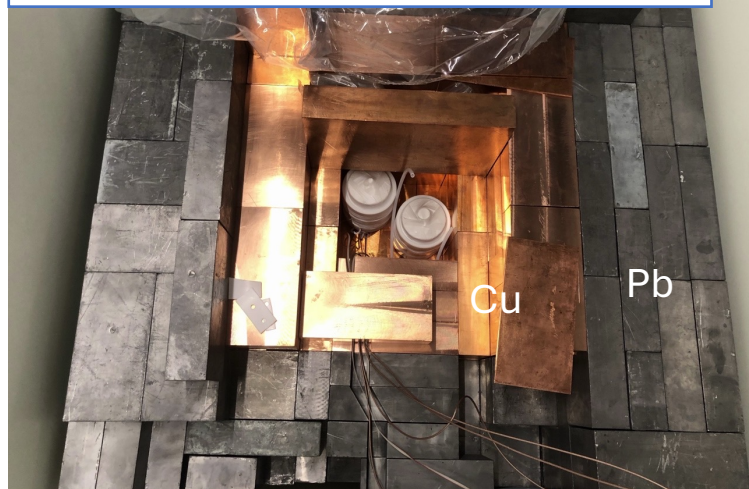
Selection efficiency is 99.7% in [0.2–3.5] MeV



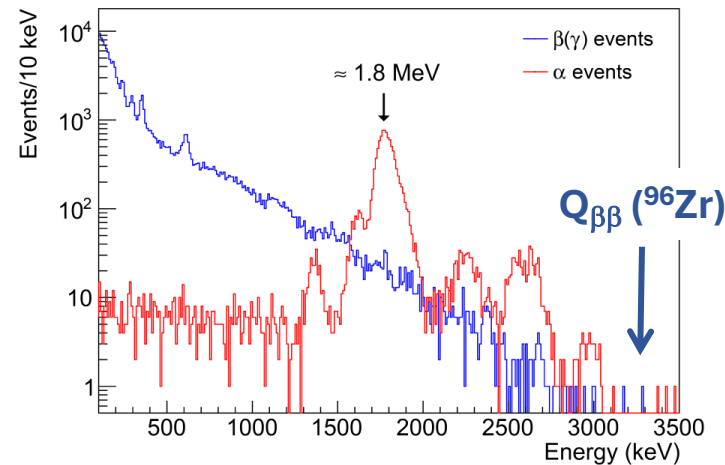
Time-amplitude analysis



DAMA/CRYS setup (2021-2022)

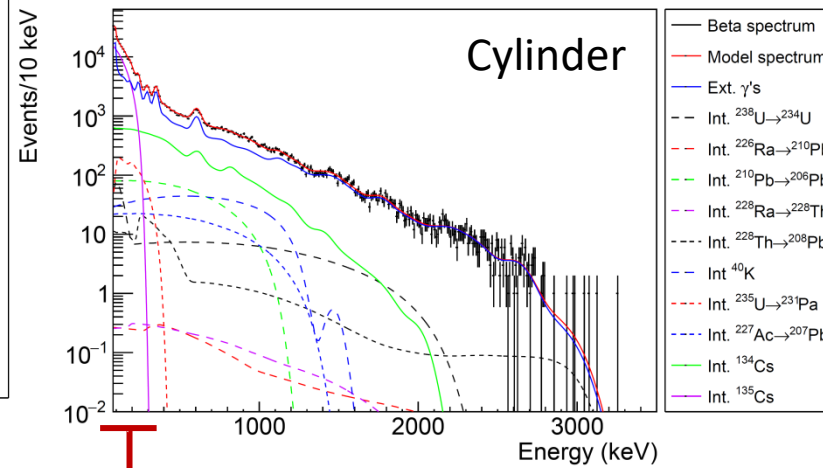
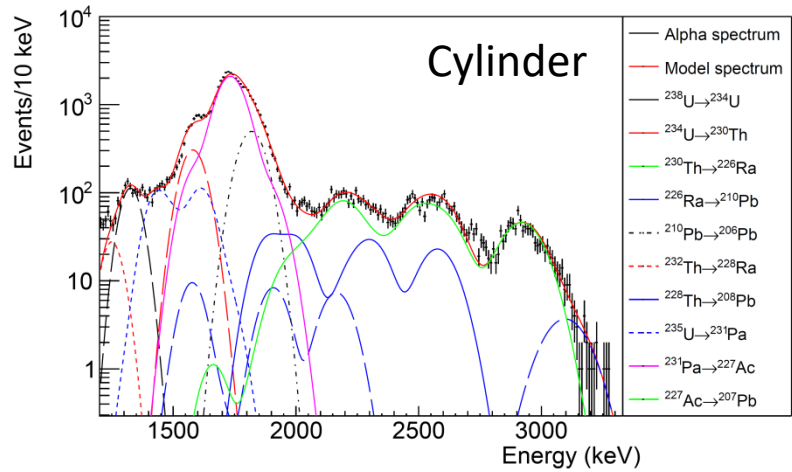
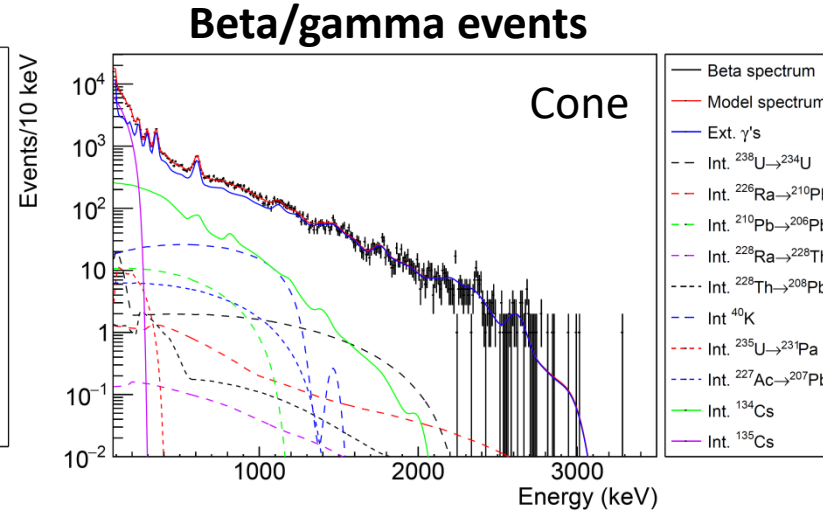
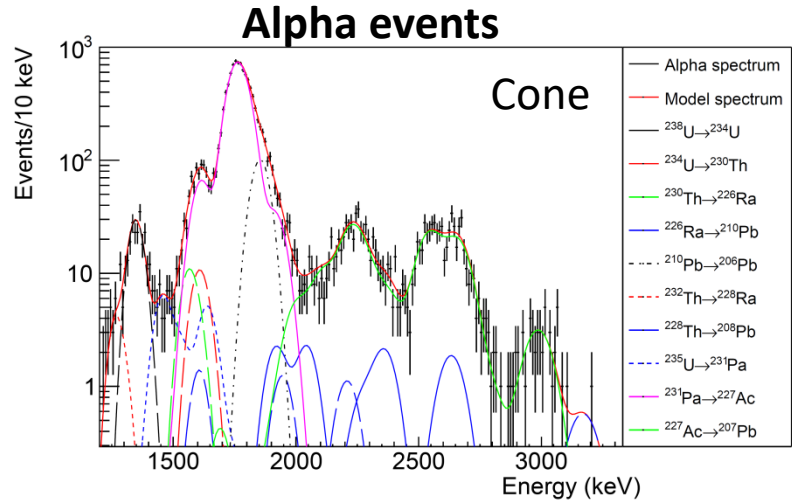


- OFHC Cu (15 cm)
- Low-activity Pb (20 cm)
- HDPE (5 cm)
- Borated HDPE (5cm)



Counting rate at ROI is 0.09 counts/(kg·keV·yr)

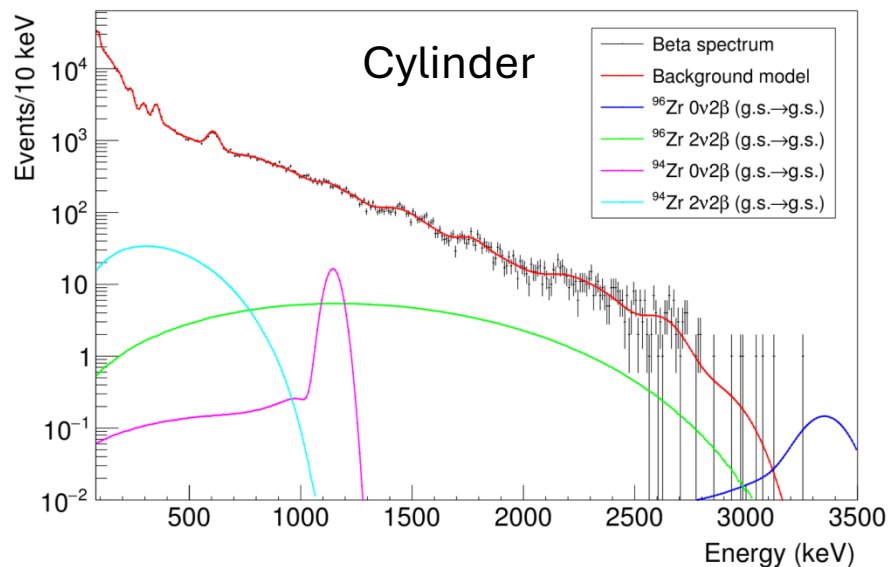
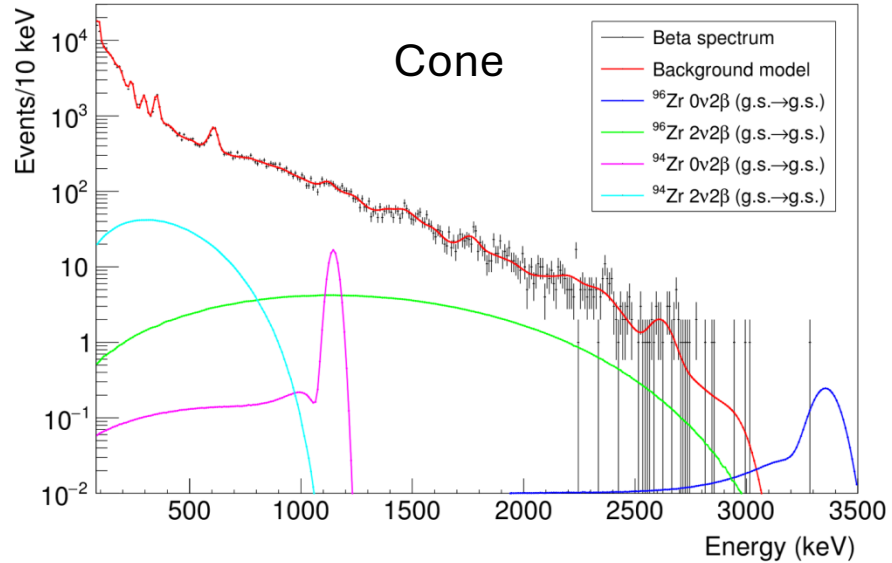
Cs₂ZrCl₆: Background model



- Comply with measurements on HPGe
- High contamination by ²³⁵U daughters
- Segregation of impurities is observed

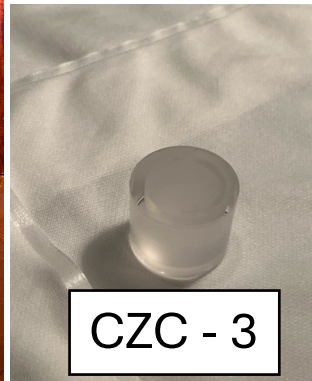
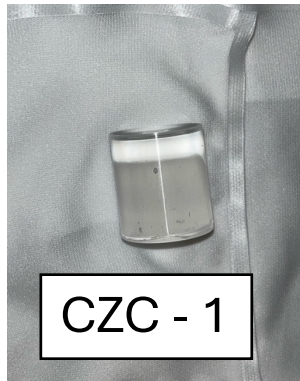
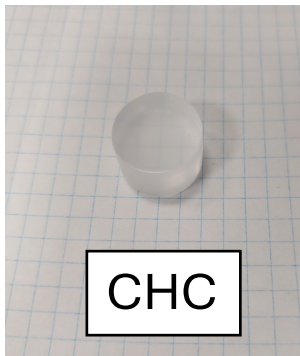
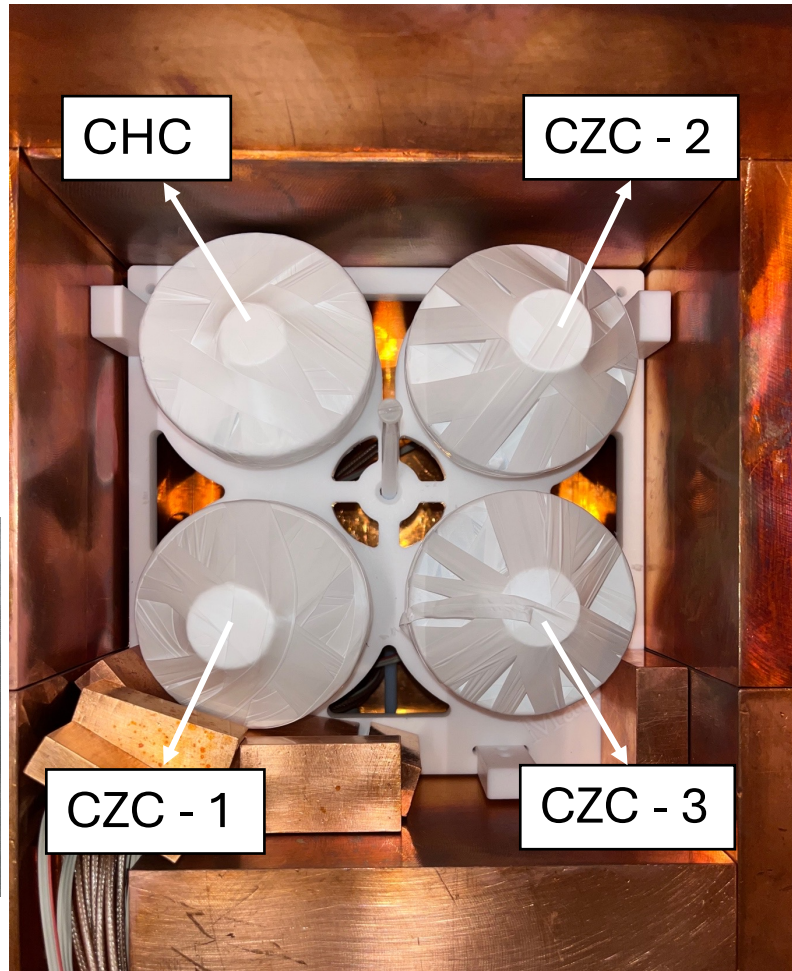
Chain	Nuclide	Internal contamination (mBq/kg)	
		Cone	Cylinder
238U	238U	0.53(4)	1.17(5)
	234U	0.2(1)	3.8(1)
	230Th	0.23(7)	< 0.02
	226Ra	0.03(3)	0.12(3)
	210Pb	2.2(2)	6.7(3)
235U	235U	0.29(4)	3.0(1)
	231Pa	21.0(3)	33.9(3)
227Ac	227Ac	0.70(3)	1.08(3)
	232Th	232Th	0.07(2)
228Th	228Th	0.05(2)	0.44(4)
	134Cs	36(4)	42(2)
	87Rb	≈ 270	≈ 290
	40K	6(1)	5(1)

Experimental limits on various decay modes in $^{94,96}\text{Zr}$ isotopes

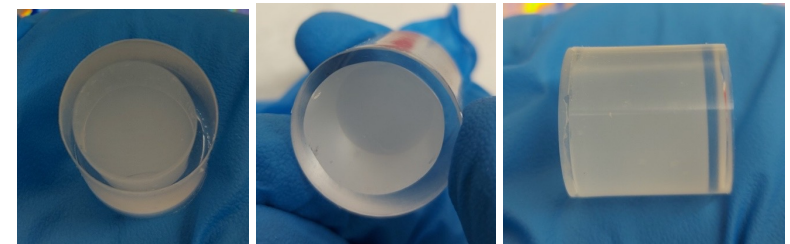


Transition	Decay mode	Final state of daughter nucleus, keV	Experimental limit on $T_{1/2}$ at 90% C.L., yr
$^{96}\text{Zr} \rightarrow ^{96}\text{Mo}$	$0\nu 2\beta$	g.s.	$> 1.5 \times 10^{20}$
		$2_1^+, 778$	$> 1.5 \times 10^{19}$
	$2\nu 2\beta$	g.s.	$> 7.4 \times 10^{17}$
		$2_1^+, 778$	$> 3.8 \times 10^{17}$
	β	g.s.	$> 1.0 \times 10^{17}$
$^{94}\text{Zr} \rightarrow ^{94}\text{Mo}$	$0\nu 2\beta$	g.s.	$> 2.6 \times 10^{19}$
		$2_1^+, 871$	$> 3.8 \times 10^{18}$
	$2\nu 2\beta$	g.s.	$> 2.4 \times 10^{18}$
		$2_1^+, 871$	$> 1.9 \times 10^{17}$

New low-background measurements in DAMA/CRYS setup (LNGS)



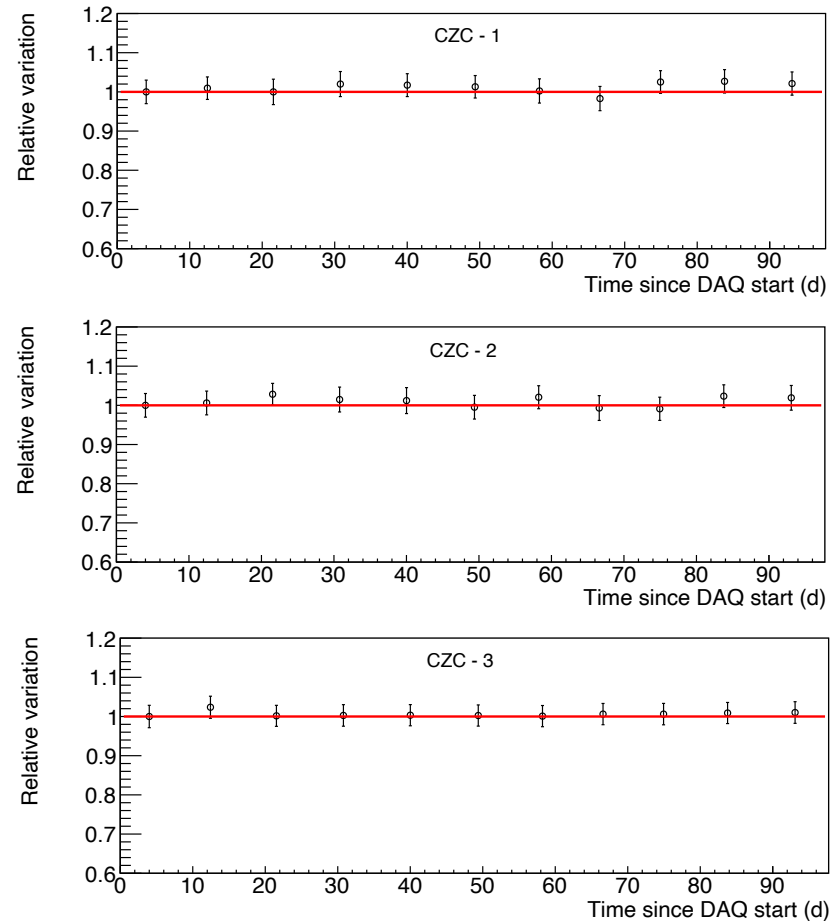
- ✓ Three new Cs_2ZrCl_6 crystals + one Cs_2HfCl_6
- ✓ Total mass of 3 CZC = 59.5 g, mass of CHC = 16.87 g.
- ✓ FWHM = 6-8% @ 662keV
- ✓ Produced from high purity and purified raw materials (> 99.99%)
- ✓ **CZC crystals are encapsulated in a silicon-based resin + quartz window**
- ✓ Modified experimental setup
- ✓ Measurements started on June 30th, 2023, for a total of 97.7 days live time



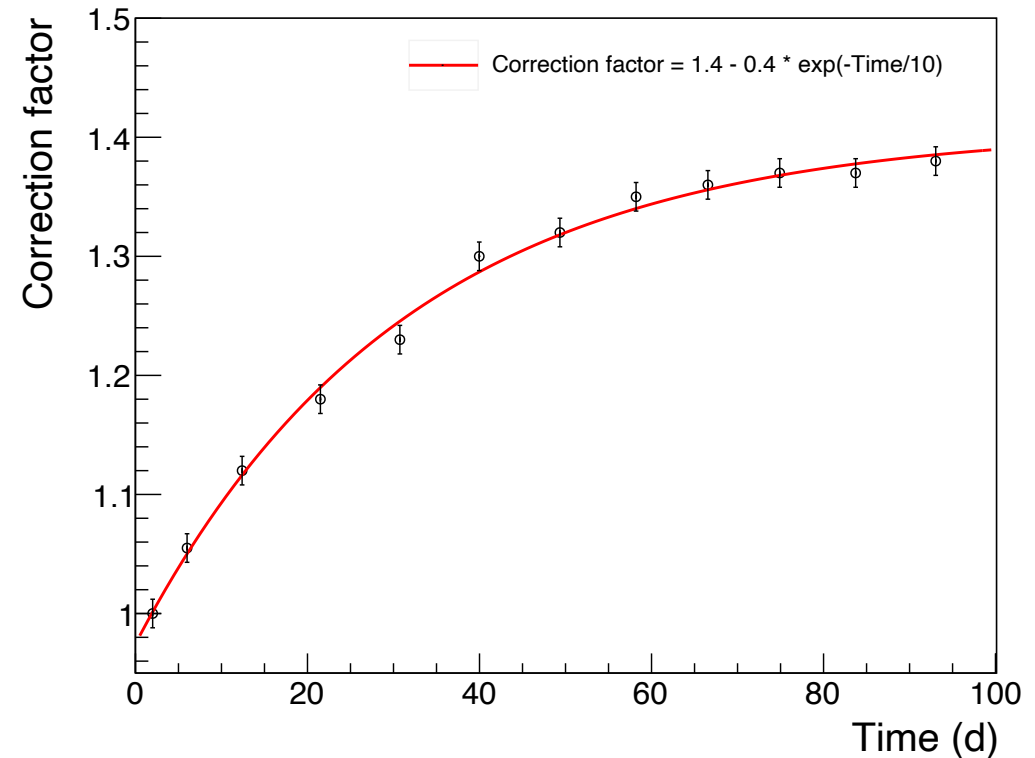
DAMA/CRYS setup at LNGS

Detectors time stability and determination of the calibration parameters

Percentage variation of the position of a reference peak during the data taking



Instead for the **CHC** a **small shift in time**: maybe due to the hygroscopicity of crystal (not encapsulated).

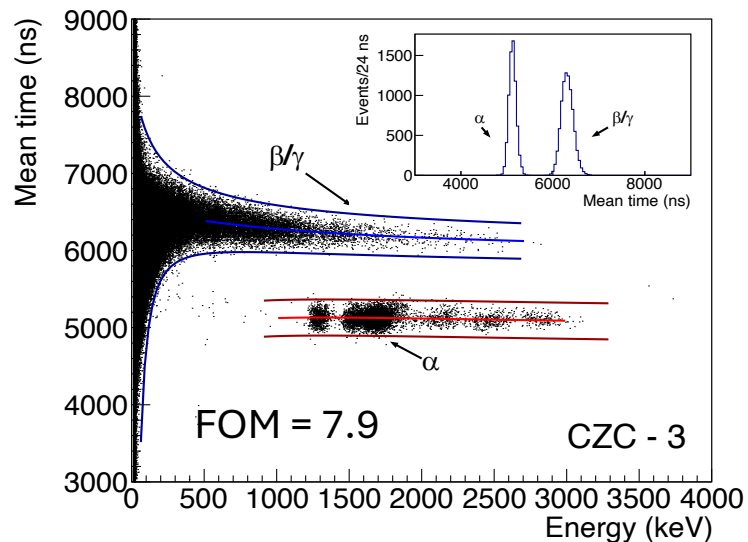
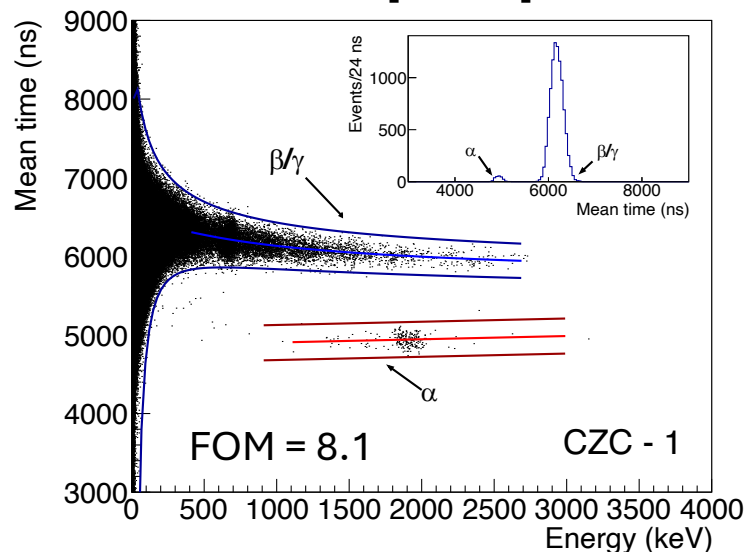


The reference peaks remain rather stable within the experimental errors, with a typical variation <math>< 1\%</math>.

Correction factor to the calibration parameters as a function of time.

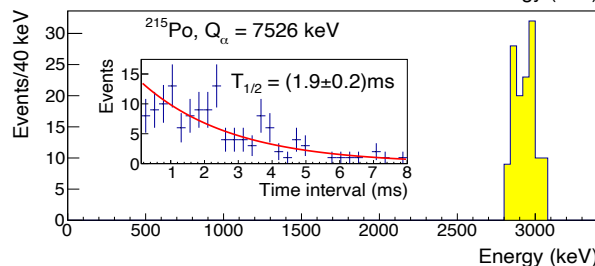
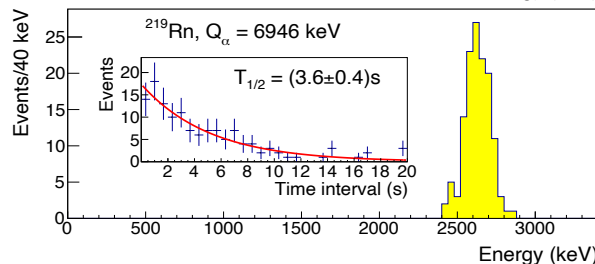
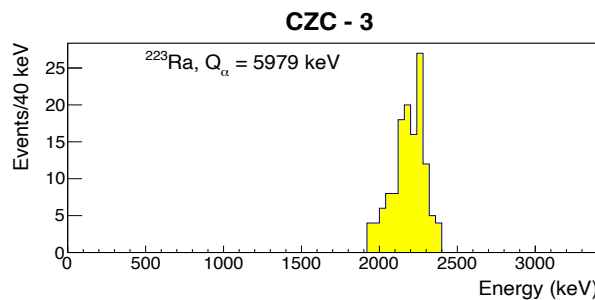
Data analysis of the Cs₂ZrCl₆ crystals

Selection efficiency is
99.7% in [0.5–3.5] MeV

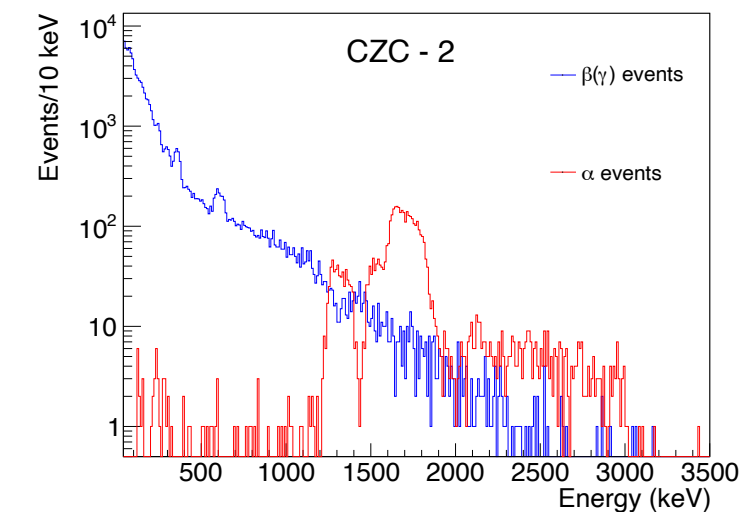
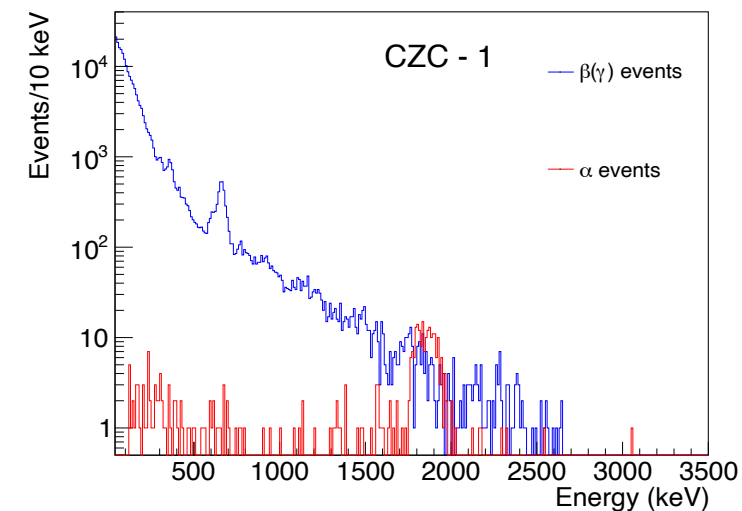


Time-amplitude analysis

A(²²⁷Ac) < 0.020 mBq/kg in CHC
< 0.017 mBq/kg in CZC - 1
= 0.56(6) mBq/kg in CZC - 2
= 0.88(8) mBq/kg in CZC - 3



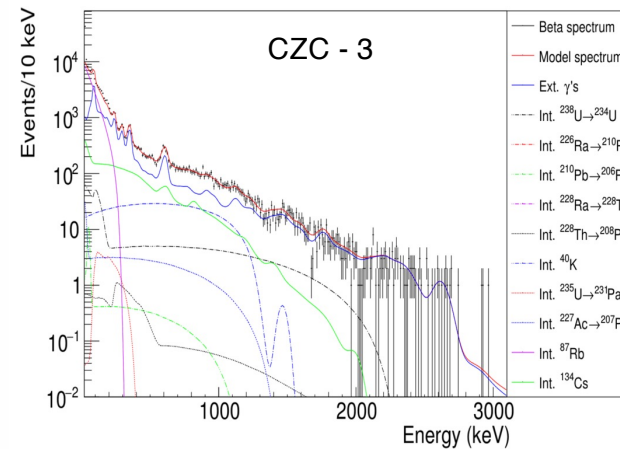
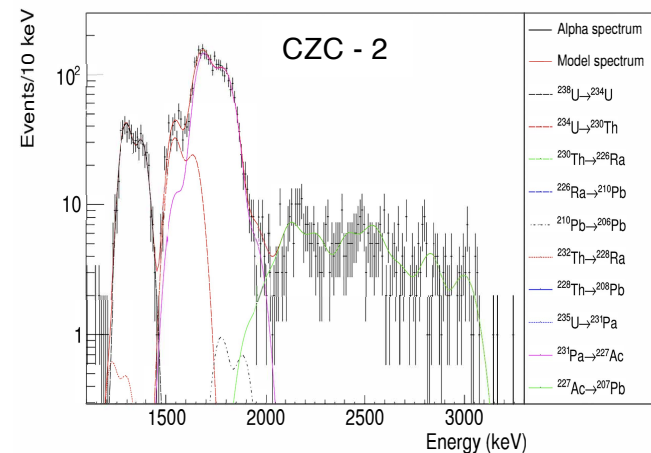
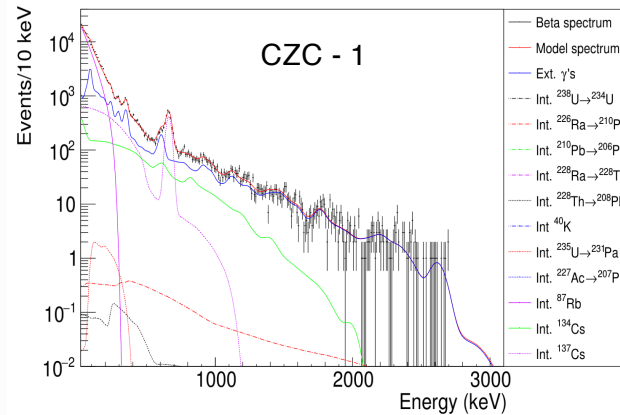
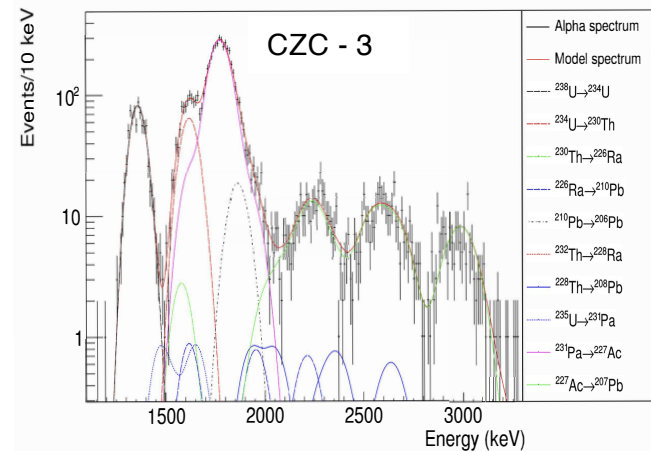
Measured energy spectra over 97.7 days of data taking



Cs₂ZrCl₆: Background model

Alpha events

Beta/gamma events



Contribution of external gammas from PMT's is dominant.

Chain	Nuclide	Internal contamination (mBq/kg)		
		CZC - 1	CZC - 2	CZC - 3
238U	238U	< 0.08	3.16(14)	4.58(18)
	234U	< 0.12	2.86(22)	4.20(33)
	230Th	< 0.12	< 0.28	< 0.6
	226Ra	< 0.05	< 0.06	< 0.17
235U	210Pb	< 1.3	< 0.6	1.32(38)
	235U	< 0.14	< 0.16	< 0.37
	231Pa	< 1.3	16.95(48)	24.69(56)
232Th	227Ac	< 0.013	0.62(3)	0.94(6)
	232Th	< 0.10	< 0.12	< 0.12
	228Th	< 0.011	< 0.04	< 0.16
	137Cs	100(3)	-	-
	134Cs	58(6)	42(7)	55(7)
	87Rb	1067(5)	318(14)	441(9)
	40K	< 1.1	11(2)	17(3)

Perspectives and conclusions

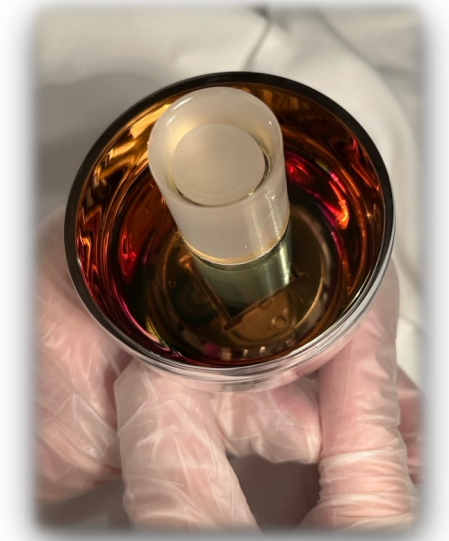
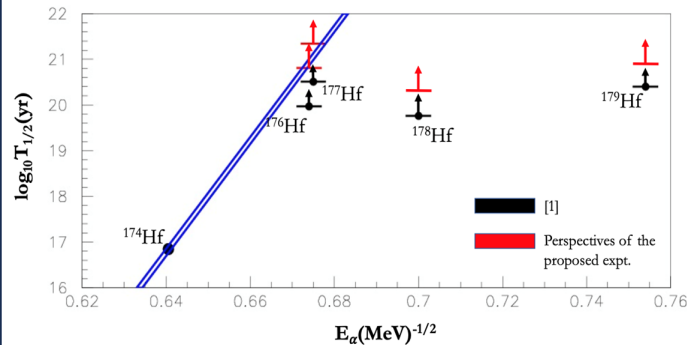


Diagram $T_{1/2}$ vs the inverse of the square root of α energy in MeV.



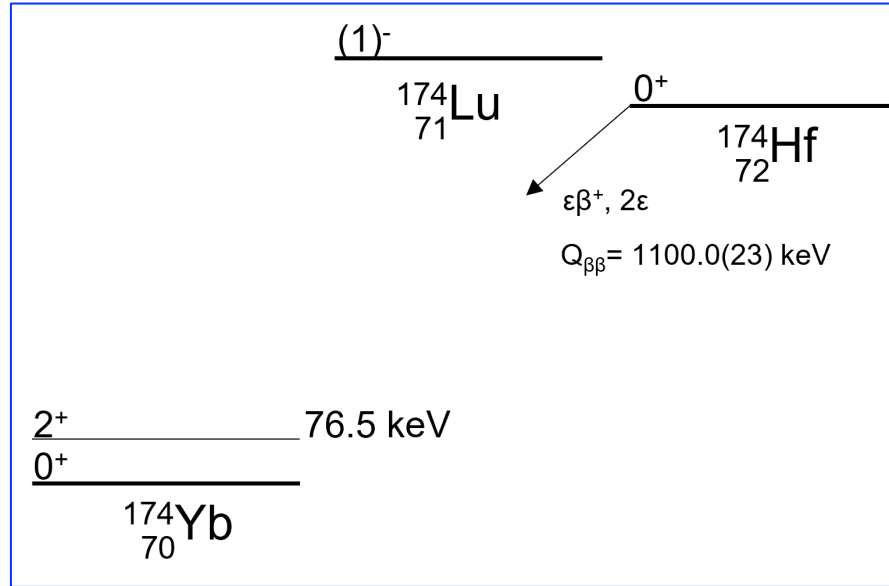
- The **blue band** is the extrapolation of the predictions on $T_{1/2}$ for all the Hf isotopes using the Geiger-Nuttall scaling law considering the data point observed in Ref. [1] *NPA 1002 (2020) 121941*.
- The **red symbols** represent the sensitivity that the measurement can reach using CHC crystal scintillators with 43.83 kg × day of total exposure.

- First experiment using a Cs_2HfCl_6 crystal scintillator in coincidence with a HP-Ge detector has observed α decay of ^{174}Hf to the g.s. with a $T_{1/2} = 7.0(1.2) \times 10^{16}$ yr [*V. Caracciolo et al. NPA 1002 (2020) 121941*].
- First two Cs_2ZrCl_6 scintillating crystals have been grown in Queen's University and studied at the LNGS, Italy to search for 2β decay of $^{94,96}\text{Zr}$ isotopes [*P. Belli et al. Eur. Phys. J. A 59, 176 (2023)*].
- New experiment using a CHC crystal scintillator and three CZC crystal scintillators has been performed in the DAMA/CRYSS setup at LNGS [*P. Belli et al 2024 JINST 19 P05037, and to appear*].
- New measurement of α decay of ^{174}Hf to the g.s.: $T_{1/2} = 3.8^{+1.7}_{-0.9} \times 10^{16}$ yr.
- A new experiment is ongoing with 4 CHC crystals ($\varnothing 26 \times 20 \text{ mm}^3$) encapsulated in silicon-based sealant.
- Compounds with a general formula A_2MX_6 , where A = Li, Na, K, Rb, Cs, Tl; M = Sn, Te, Hf, Zr, Pt, Os, Re, Ru; and X = Cl, Br, or I, are flexible to the element of interest that can be embedded for its fundamental studies.

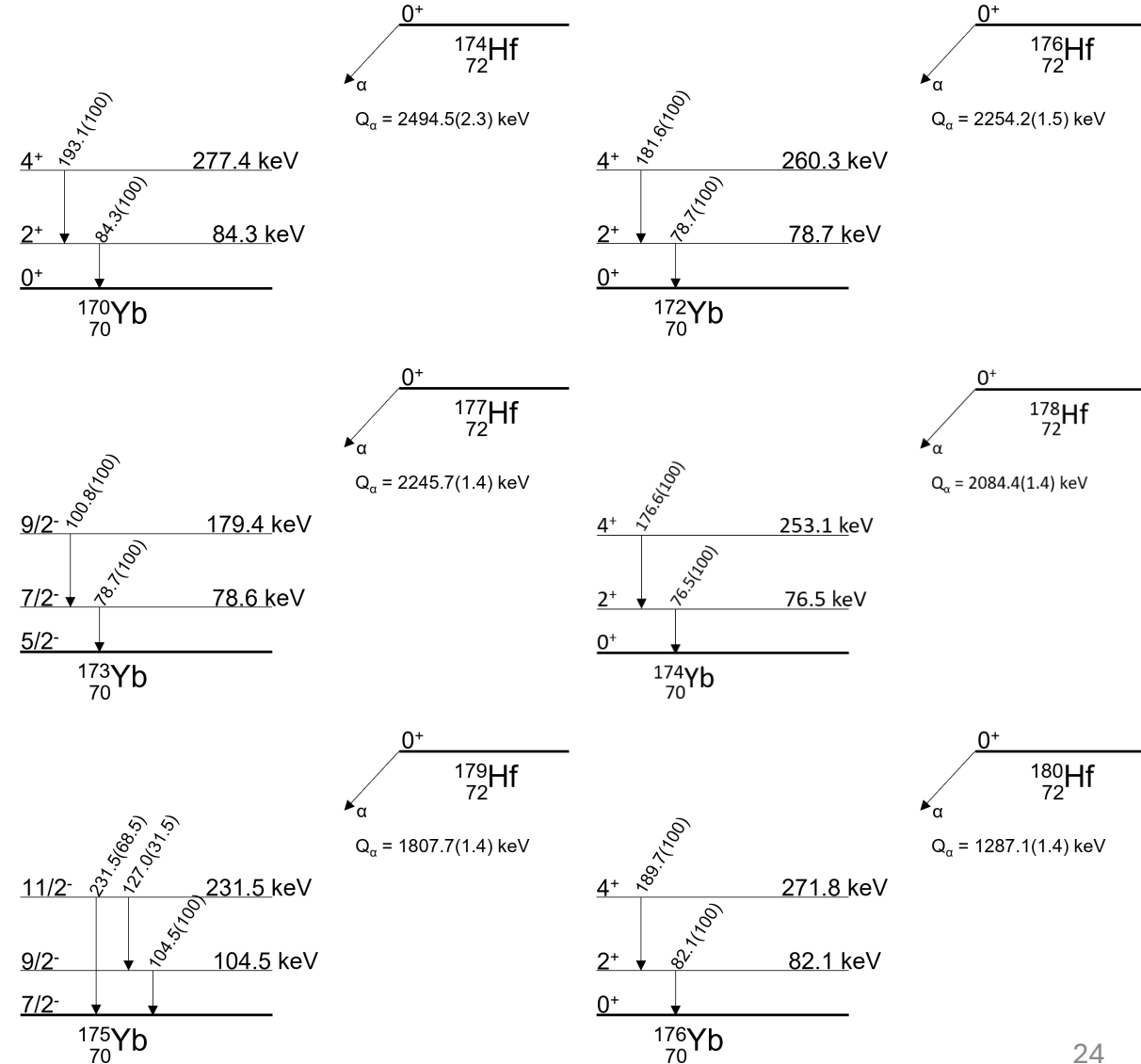
BACKUP SLIDES

Simplified decay schemes of naturally occurring Hf isotopes

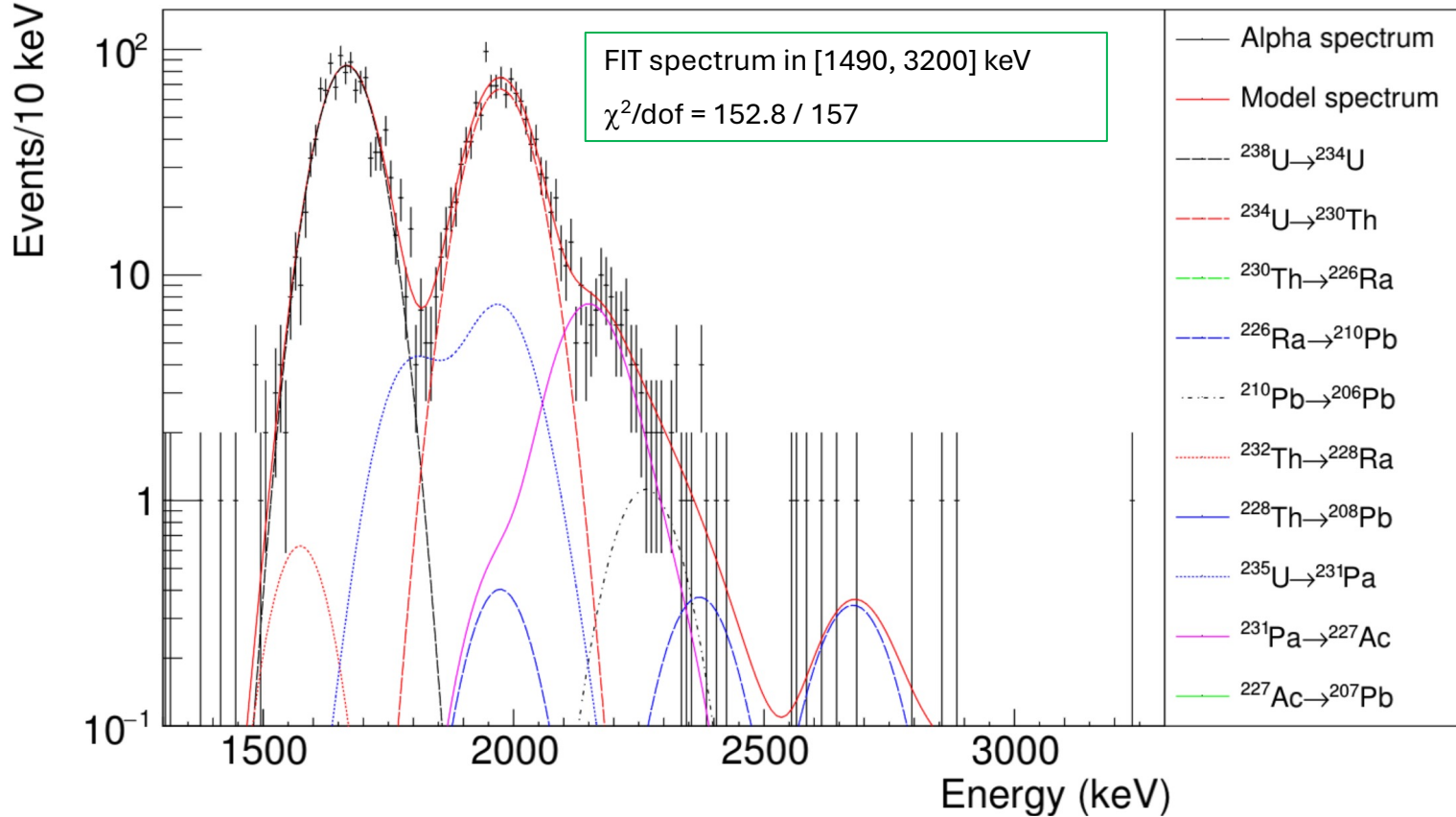
2ε, εβ⁺ decay of ¹⁷⁴Hf



α decays of Hf isotopes considering the first two excited energy levels of the daughter nuclei. Energies of the excited levels and of the emitted γ quanta are shown. Relative probabilities of a single energy level are given in parentheses. The ¹⁷⁵Yb isotope decays via β⁻ with T_{1/2} = 4.185(1) d, while all the other Yb nuclei are stable.



Cs₂HfCl₆: background model of the α spectrum



$$\alpha/\beta = 0.2777(65) + 0.0278(14) \cdot E_{\alpha}[\text{MeV}]$$

$$\text{FWHM}(\text{keVee}) = \sqrt{5.97 * E[\text{keVee}] + 0.0014 \cdot E^2[\text{keVee}]}$$

Chain	Nuclide	Internal contamination (mBq/kg)
		CHC (16.87 g)
^{238}U	^{238}U	7.6(3)
	^{234}U	6.7(5)
	^{230}Th	< 0.5
	^{226}Ra	0.04(2)
	^{210}Pb	0.12(7)
^{235}U	^{235}U	1.3(5)
	^{231}Pa	0.92(13)
	^{227}Ac	<0.005
^{232}Th	^{232}Th	< 0.22
	^{228}Th	<0.02
	^{147}Sm	0.25(10)
	^{134}Cs	44(8)
	^{87}Rb	< 400
	^{40}K	< 2.3

Half-life of α decay of ^{174}Hf to the g.s. of ^{174}Yb

Area of 2nd peak = $118 \pm 11(\text{stat}) \pm 35(\text{sys}) = 118 \pm 37$

Half-life:

$$T_{1/2} = \ln 2 \cdot N \cdot \epsilon \cdot t / S$$

- N (number of nuclides) = $\frac{M}{W} \cdot \delta \cdot N_A = 2.412 \times 10^{19}$
- ϵ is the PSD efficiency which corresponds to 99%;
- t is the measurement time (= 2344.8 h = 0.26767 yr);

$M = 16.87$ g;

$W(\text{Cs}_2\text{HfCl}_6) = 657$ g/mole;

$\delta(^{174}\text{Hf}) = 0.156(6)$ %

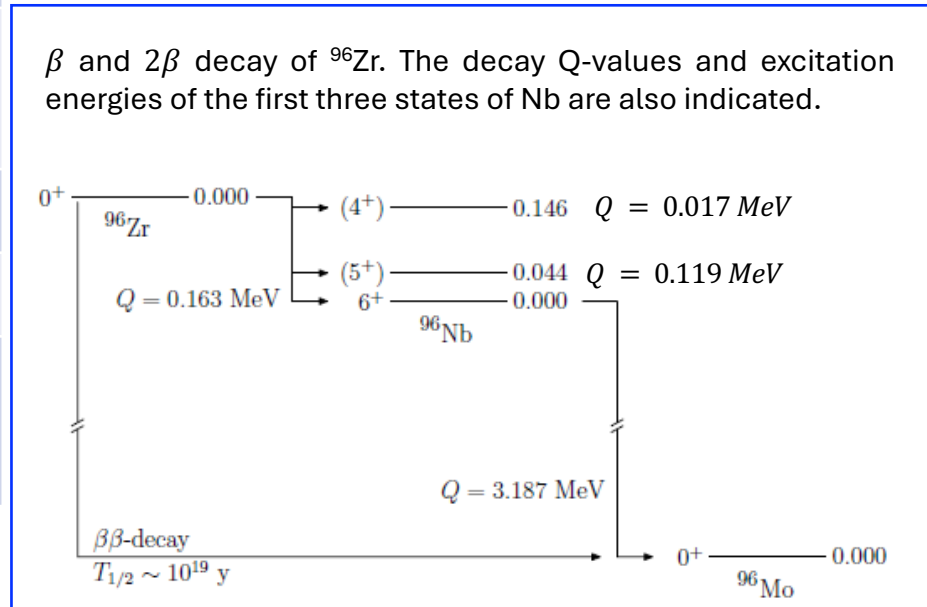
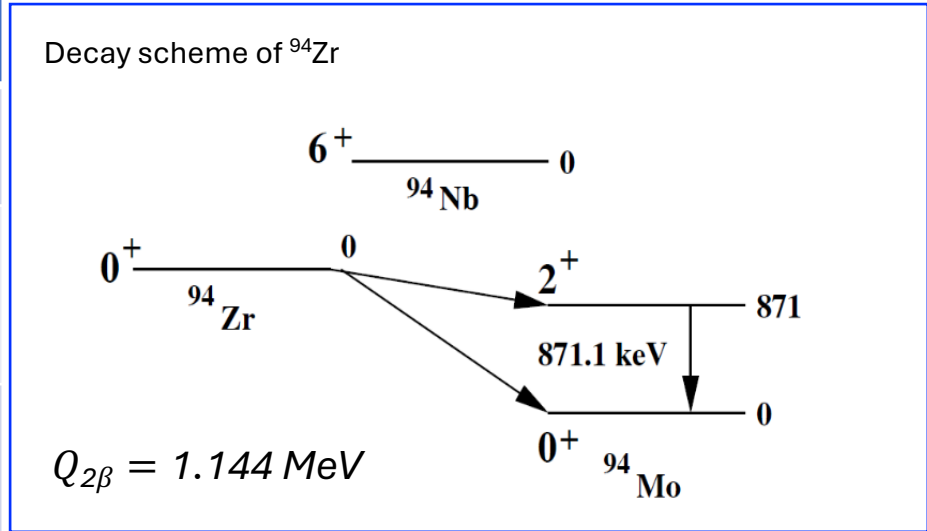
$\Rightarrow T_{1/2} = [3.8_{-0.3}^{+0.4}(\text{stat})_{-0.9}^{+1.6}(\text{sys})] \times 10^{16} = 3.8_{-0.9}^{+1.7} \times 10^{16}$ yr of α decay of ^{174}Hf

Comparing with result in [*NPA 1002 (2020) 121941*]: $\frac{|3.8-7.0|}{\sqrt{(1.7)^2 + (1.2)^2}} = 1.5$

Theoretical predictions: $(3.5 - 7.4) \times 10^{16}$ yr.

Search for 2β decay in $^{94,96}\text{Zr}$ and for ^{96}Zr 's β decay

Experiment	Transition	$T_{1/2}$ 90% C.L. (y)	Technique	Ref.
ZICOS, (Kamioka Observatory, Japan)	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 0^+_1$ (g.s.)	under construction	Organic liquid scintillator	[1]
NEMO I, II, III, Frejus (France) (next: SuperNEMO)	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 0^+_1$ (g.s.)	$>9.2 \times 10^{21}$	Tracker detector	[2]
		$>1.29 \times 10^{22}$		[3]
Kimballton Underground Research Facility, (USA)	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 2^+_1$	$>3.1 \times 10^{20}$	HP-Ge	[4]
Collaboration at Fréjus Underground Laboratory	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 2^+_1, 0^+_1, 2^+_2, 2^+_3$	$>(2.6 - 7.9) \times 10^{19}$	HP-Ge	[5]
Collaboration at LNGS	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 2^+_1$	$>3.8 \times 10^{19}$	HP-Ge	[6]
TILES (TIFR, Mumbai)	$^{94}\text{Zr } 0^+ \rightarrow ^{94}\text{Mo } 2^+_1$	$>5.2 \times 10^{19}$	HP-Ge	[7]
Kimballton Underground Research Facility, (USA)	$^{96}\text{Zr } 0^+ \rightarrow ^{96}\text{Mo } 6^+$	$>2.4 \times 10^{19}$	HP-Ge	[8]



[1] EPS-HEP (2019) 437

[2] NPA 847 (2010) 168

[3] PhD U. Coll. London (2015)

[4] S.W. Finch et W. Tornow, Phys. Rev. C 92 (2015) 045501

[5] J. Phys. G: Nucl. Part. Phys. 22 (1996) 487

[6] C. Arpesella et al. Lett. 27 (l) (1994) pp. 29–34

[7] N. Dokania et al. J. Phys. G: Nucl. Part. Phys. 45 (2018) 075104

[8] S.W. Finch, W. Tornow, Nucl. Inst. Meth. A 806(2016)70–74

[9] J. Heeck and W. Rodejohann 2013 EPL 103 32001