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Low level measurements for rare nuclear transitions in Hf isotopes

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LNGS 2022
ICRM - LLRMT

- **Passive source approach** (source≠detector): gamma-ray spectrometry technique
- **Active source approach** (source=detector) : crystal scintillator

Both the techniques are implemented and optimized to investigate rare nuclear decays in Hf nuclides, such as α decay to the ground level and to the first excited state and 2β decays of ^{174}Hf.

Interesting in studying the 34 $2\beta^+$ emitters

- **2β** decay without the presence of neutrinos, if observed, could open a new window beyond the Standard Model.
- To **test calculations** of **different nuclear shapes** and the **decay modes** that involve the vector and **axial-vector weak effective coupling constants**; possible study of the "resonant effect" on the 0ν2ε mode;
- The nuclear matrix elements for the **2ν** mode and for the **0ν mode** can be **related** to each other through relevant parameters: in the free nucleon interaction, **the g_A value is 1.2701**, but, when considering a nuclear decay, there are indications that the phenomenological axial-vector coupling value is reduced at **$g_A < 1$**, more precisely: **$g_A \approx 1.269\ A^{-0.18}$ or $g_A \approx 1.269\ A^{-0.12}$**, depending on the nuclear model adopted to infer the g_A value.

<u>2β investigation with various nuclei would shed new light in constraining these and other important model-dependent parameters.</u>

$$2\nu 2\beta^+ :\ {}^A_Z X \rightarrow {}^A_{Z-2} X + 2e^+ + 2\nu_e$$

L conserved

$$0\nu 2\beta^+ :\ {}^A_Z X \rightarrow {}^A_{Z-2} X + 2e^+$$

L violated ($\Delta L = 2$) → massive Majorana neutrino

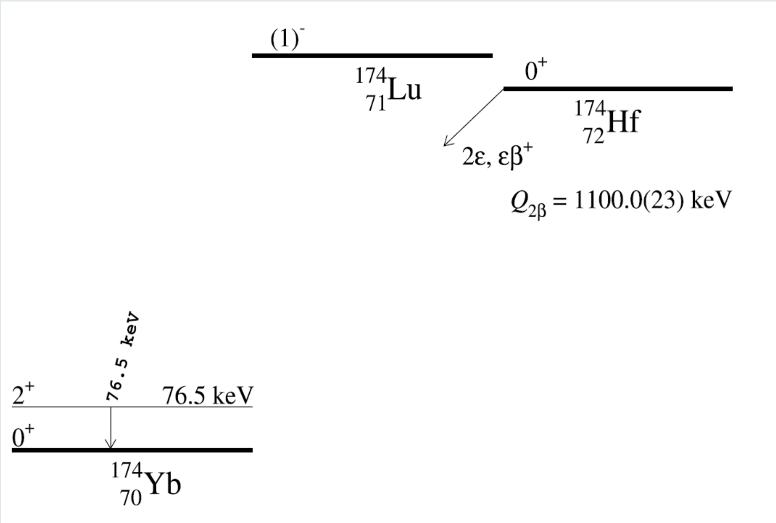
$$0\nu\epsilon\beta^+ :\ e^- + {}^A_Z X \rightarrow {}^A_{Z-2} X + e^+ + \text{X-rays}$$

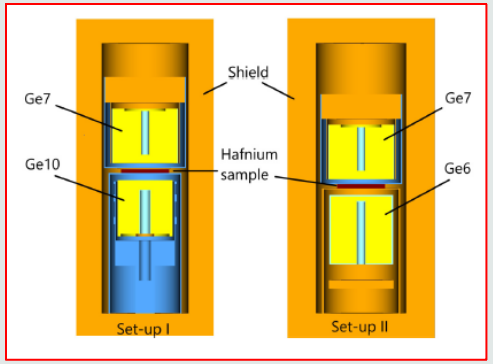
$$0\nu 2\epsilon :\ e^- + e^- + {}^A_Z X \rightarrow {}^A_{Z-2} X^* \rightarrow {}^A_{Z-2} X + \gamma + \text{X-rays}$$

Search for **2β** decay in Hf isotopes using passive source approach

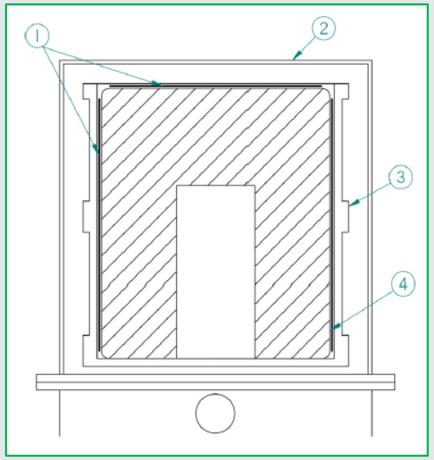
HPGe-detector

The isotope ^{174}Hf is one of the potentially 2ε, $\varepsilon\beta^+$ radioactive nuclides with the energy of decay $Q_{2\beta}$=1100.0(23) keV and the isotopic abundance δ=0.156(6)%.





m_{Hf} = 179.8 g,
∅=59.0 mm × 5.0mm,
225 m underground:
HADES lab of Joint Research Centre of European Commission (Geel, Belgium).
75 days.



Hf foil: 0.25(1) mm thick,
m_{Hf} =55.379(1) g,
Located underground at **LNGS**,
310 days.

Section view of the detector and sample (not to scale) with 1) hafnium foils on the top and wrapping the Ge crystal acting as the target and high-voltage contact,
2) copper end cap of 1mm thickness,
3) copper HP-Ge crystal holder, and
4) HP-Ge semi-coaxial p-type crystal.

NPA 996 (2020) 121703

NPA 1012 (2021) 122212

Channel of the decay	Decay Mode	Level of daughter nucleus J^π, energy (keV)	Experimental limit of $T_{1/2}$ (90% C.L.) (y) [26]	[27]
$2K$	2ν	g.s.	$\geq 7.1 \times 10^{16}$	$\geq 1.4 \times 10^{16}$
KL	2ν	g.s.	$\geq 4.2 \times 10^{16}$	$\geq 1.4 \times 10^{16}$
$2K$	2ν	2^+, 76.5	$\geq 5.9 \times 10^{16}$	$\geq 7.9 \times 10^{16}$
KL	2ν	2^+, 76.5	$\geq 3.5 \times 10^{16}$	$\geq 7.9 \times 10^{16}$
$2L$	2ν	2^+, 76.5	$\geq 3.9 \times 10^{16}$	$\geq 7.9 \times 10^{16}$
$2K$	0ν	g.s.	$\geq 5.8 \times 10^{17}$	$\geq 2.7 \times 10^{18}$
KL	0ν	g.s.	$\geq 1.9 \times 10^{18}$	$\geq 4.2 \times 10^{17}$
$2L$	0ν	g.s.	$\geq 7.8 \times 10^{17}$	$\geq 3.6 \times 10^{17}$
$2K$	0ν	2^+, 76.5	$\geq 7.1 \times 10^{17}$	$\geq 2.4 \times 10^{18}$
KL	0ν	2^+, 76.5	$\geq 6.2 \times 10^{17}$	$\geq 3.1 \times 10^{17}$
$2L$	0ν	2^+, 76.5	$\geq 7.2 \times 10^{17}$	$\geq 9.4 \times 10^{17}$
$K\beta^+$	$2\nu + 0\nu$	g.s.	$\geq 1.4 \times 10^{17}$	$\geq 5.6 \times 10^{16}$
$L\beta^+$	$2\nu + 0\nu$	g.s.	$\geq 1.4 \times 10^{17}$	$\geq 5.6 \times 10^{16}$

Interesting in studying rare α decay

Various **theoretical models are continuously developed or improved**, e.g., motivated by searches for stable or long-lived superheavy isotopes and predictions of their half-lives.

The study on the **nuclear instability offers details** about the **nuclear structure**, the nuclear levels and the properties of nuclei.

The phenomenon of α decay can offer information about the **fusion-fission reactions** since the α decay process involves sub-barrier penetration caused by the interaction between the α particle and the nucleus.

Understanding the nuclear properties is essential also for **nuclear and particle astrophysics studies**, for example, α–capture reactions (equivalent to the inverse α–decay process) are important for nucleosynthesis and β–delayed fission, together with other fission modes, determine the so-called "fission recycling" in the r-process nucleosynthesis.

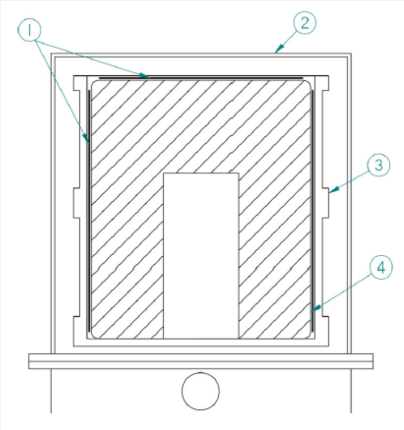
As byproduct: developments of new detectors, e.g., new crystal scintillators containing α emitters.

Search for α decay in Hf isotopes using passive source approach

Search for α decays of the naturally occurring hafnium isotopes to the **first excited levels** of the daughter nuclei. → **de-excitation γ quanta are emitted, which can be searched for by low-background γ -ray spectrometry.**

NPA 1012 (2021) 122212

Decay Isotope	E_γ [keV] (y [%])	Experimental $T_{1/2}$ [a] This work
^{174}Hf	84.3 (13.7)	$\geq 2.8 \times 10^{16}$
^{176}Hf	78.7 (10.6)	$\geq 2.7 \times 10^{17}$
^{177}Hf	78.6 (12.5)	$\geq 1.1 \times 10^{18}$
^{178}Hf	76.5 (9.6)	$\geq 1.3 \times 10^{18}$
^{179}Hf	104.5 (26.7)	$\geq 2.7 \times 10^{18}$
^{180}Hf	82.1 (12.4)	$\geq 4.6 \times 10^{17}$

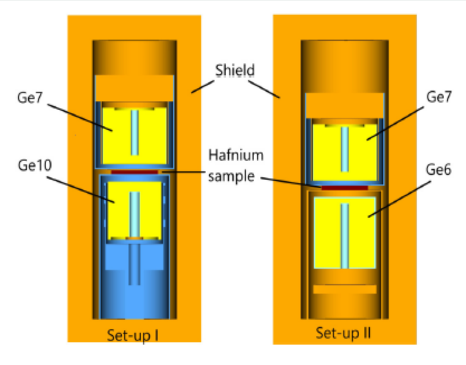


LNGS

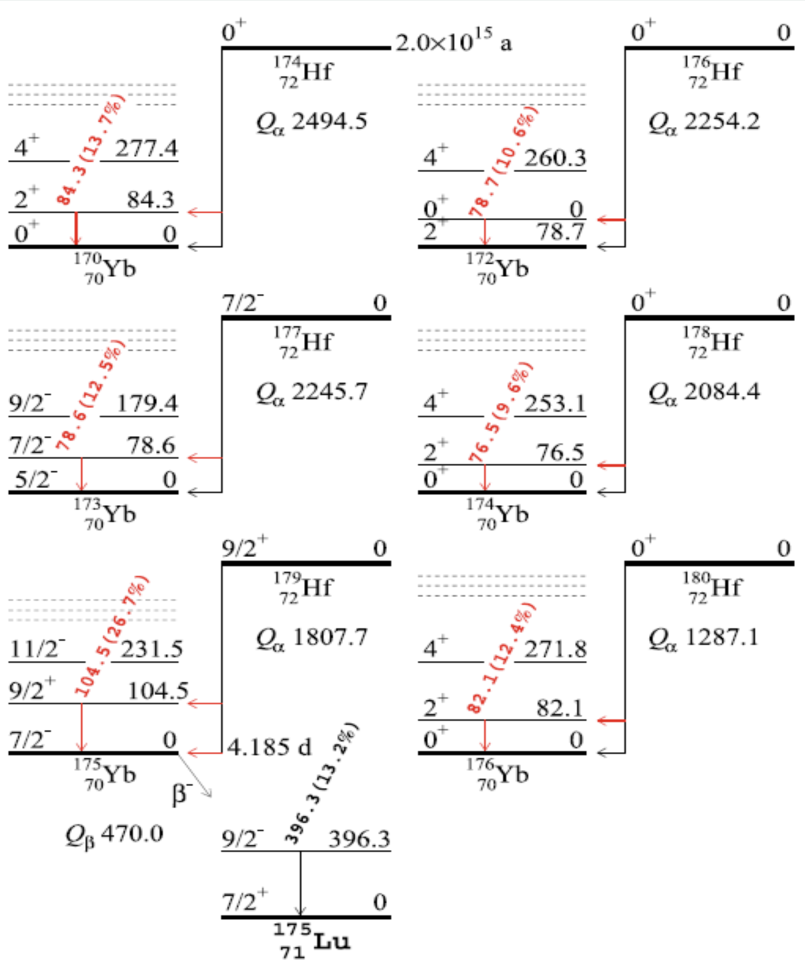
Eur. Phys. J. A (2020) 56:5

T.P. Kohman, Phys. Rev. 121, 1758 (1961)

Transition	Level of the daughter nucleus (keV)	Experimental $T_{1/2}$ (a)	Theoretical $T_{1/2}$ (a)		
			[8]	[9, 10]	[11]
^{174}Hf → ^{170}Yb	0^+, g.s.	$= 2.0(4) \times 10^{15}$ [5]	7.4×10^{16}	3.5×10^{16}	3.5×10^{16}
	2^+, 84.3	$\geq 3.3 \times 10^{15}$	3.0×10^{18}	1.3×10^{18}	6.6×10^{17}
^{176}Hf → ^{172}Yb	2^+, 78.7	$\geq 3.0 \times 10^{17}$	3.5×10^{22}	1.3×10^{22}	4.9×10^{21}
^{177}Hf → ^{173}Yb	$7/2^-$, 78.6	$\geq 1.3 \times 10^{18}$	1.2×10^{24}	9.1×10^{21}	3.6×10^{23}
^{178}Hf → ^{174}Yb	2^+, 76.5	$\geq 2.0 \times 10^{17}$	8.1×10^{25}	2.4×0^{25}	7.1×10^{24}
^{179}Hf → ^{175}Yb	$(7/2^-)$, g.s.	$\geq 2.2 \times 10^{18}$	4.0×10^{32}	4.4×10^{29}	4.7×10^{31}
	$(9/2)^+$, 104.5	$\geq 2.2 \times 10^{18}$	2.5×10^{35}	2.0×10^{32}	2.2×10^{34}
^{180}Hf → ^{176}Yb	2^+, 82.1	$\geq 1.0 \times 10^{18}$	4.1×10^{50}	4.0×10^{49}	2.1×10^{48}



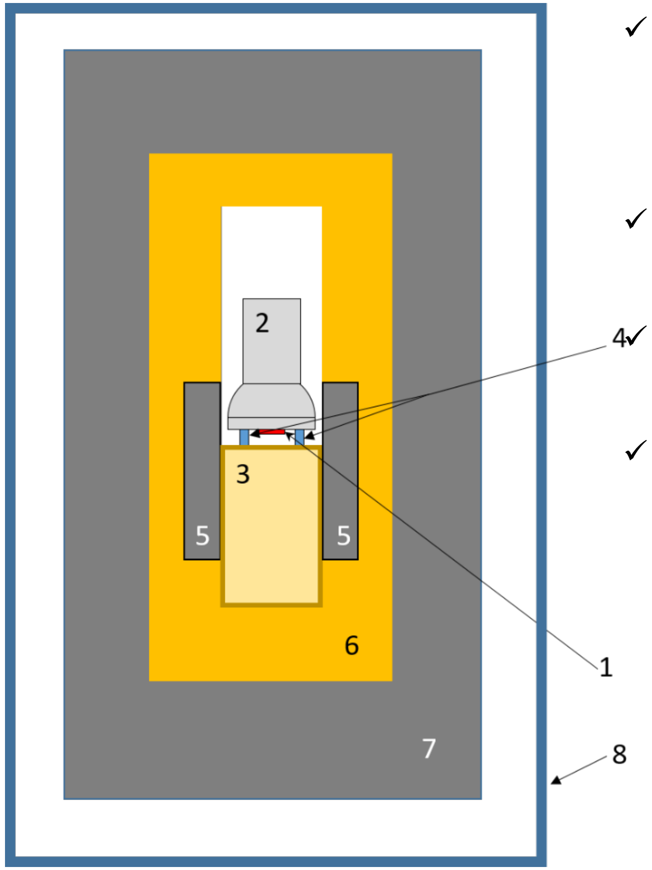
HADES laboratory (Belgium)



Search for α decay in Hf isotopes using active source approach

NPA 1002 (2020) 121941

Cs_2HfCl_6 crystal (CHC) *(in collab. with Queen's University)*

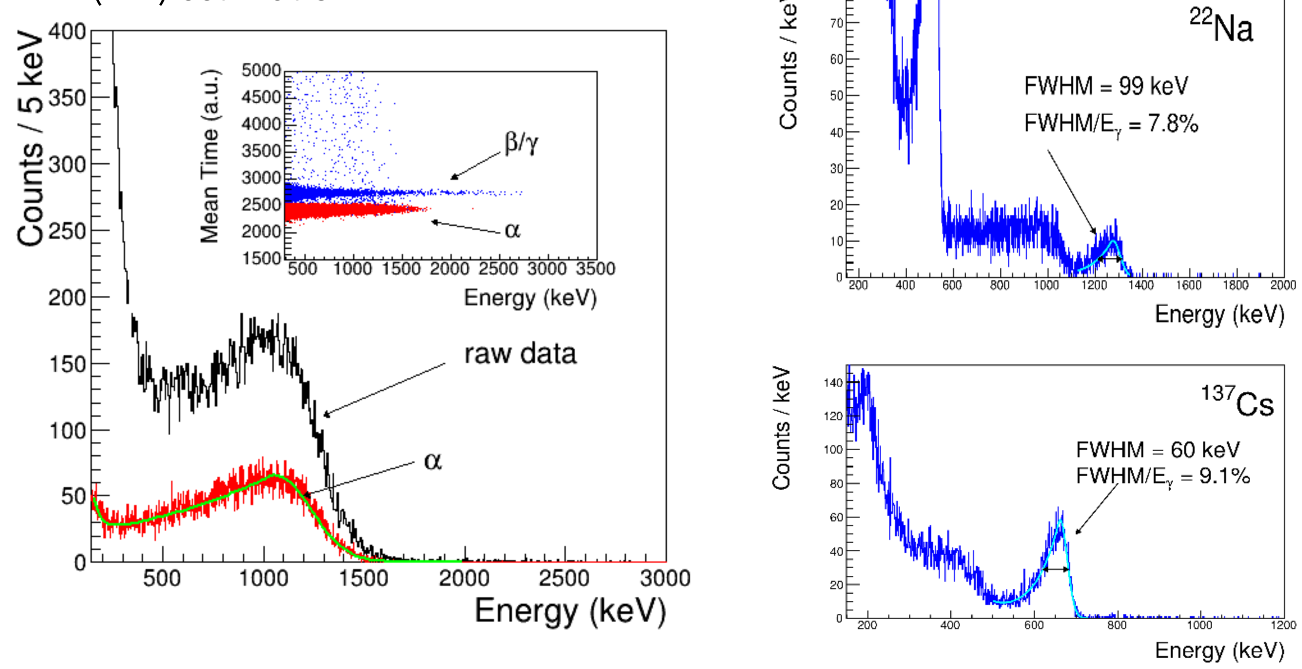


- ✓ CHC crystal (6.90(1) g) coupled low-radioactivity PMT (Hamamatsu R6233MOD)
- ✓ placed above the end-cap of the ultra-low background HP-Ge
- ✓ CAEN DT5720B digitizer 250 MSamples/s;
- ✓ 2848 h data taking

STELLA facility of the LNGS

Energy Calibration

Preliminary quenching factor (QF) estimation → ^{241}Am



Schematic cross-sectional view of the experimental set-up (not in scale). There are shown the CHC crystal scintillator (1) coupled with a 3 inches PMT (2), the HP-Ge detector (3), which is separated by a cylindrical Teflon ring (4). They are completely surrounded by a passive shield made by archaeological Roman lead (~ 2.5 cm) (5), high purity copper (~ 5 cm) (6), low radioactive lead (~ 25 cm) (7). The whole set-up (with the exception of the cold finger for the HP-Ge detector) is enclosed in a Plexiglas box (8) continuously flushed with HP-N_2 gas.

The CHC crystal scintillator

Some general properties of CHC crystal scintillators.

Effective atomic number	58
Density (g/cm^3)	3.9
Melting point (°C)	820
Crystal structure	cubic
Wavelength of emission (nm)	400–430
Average decay time (µs)	4–5

It represents one of the promising new scintillating materials for γ spectroscopy also in the field of low-level measurements:

- light output of more than 50000 photons/MeV;
- high energy resolution;
- excellent ability for pulse shape discrimination (PSD) between $\boldsymbol{\gamma}(\boldsymbol{\beta})$ and $\boldsymbol{\alpha}$ particles;
- It is also the first scintillating material containing a high fraction of Hf (~ 27% in mass) that can be easily produced using the Bridgman growing technique.

Low background measurements of the CHC crystal

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Isotopic composition of ^{nat}Hf measured in a sample of the CHC crystal by ICP-MS

Isotope	Abundance (%)
^{174}Hf	0.156(6)
^{176}Hf	5.18(5)
^{177}Hf	18.5(1)
^{178}Hf	27.2(1)
^{179}Hf	13.9(1)
^{180}Hf	35.2(2)

Concentrations of trace contaminants in the CHC crystal as measured by ICP-MS analysis. The limits are at 68% C.L.

Nuclide	Concentration (ppb)
^{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

Nuclide	Q_α (keV) [10]	$T_{1/2}$ (y) [10]	Isotopic Abundance (%) [2]	E_α (keV)	**Expected Counts in the ROI (see later)**
^{144}Nd	1906.4(17)	$2.29(16)\times10^{15}$	23.798(19)	1854.8(17)	<0.007
^{147}Sm	2311.2(10)	$1.060(11)\times10^{11}$	15.00(14)	2249.9(10)	36(6)
^{148}Sm	1986.9(10)	$7(3)\times10^{15}$	11.25(9)	1934.6(10)	$3.6(1)\times10^{-4}$
^{152}Gd	2204.4(10) [11]	$1.08(8)\times10^{14}$	0.20(3)	2147.8(10)	$<1\times10^{-3}$
^{186}Os	2820.4(13)	$2.0(11)\times10^{15}$	1.59(64)	2761.0(13)	$<6\times10^{-4}$
^{190}Pt	3252.6(6)	$6.5(3)\times10^{11}$	0.012(2)	3185.5(6)	<0.1
^{209}Bi	3137.3(8)	$2.01(8)\times10^{19}$	100	3078.4(8)	$<4\times10^{-7}$

Low background measurements of the CHC crystal

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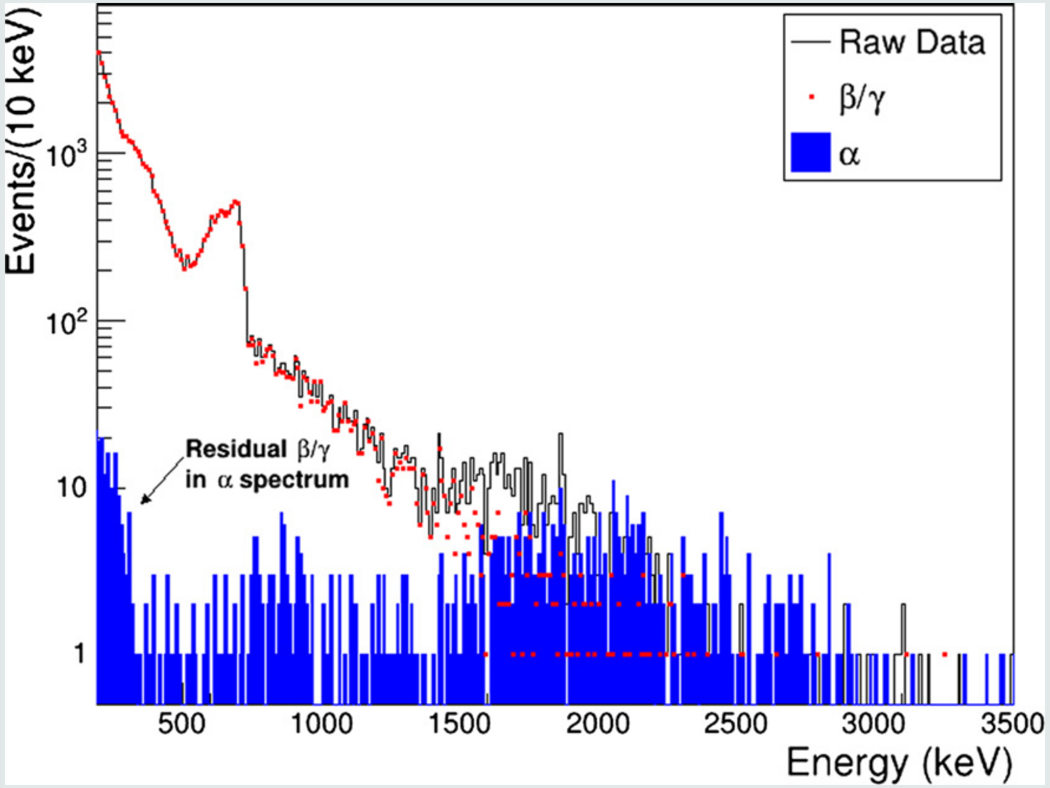
Chain	Nuclide	Activity (mBq/kg)
	^{40}K	$0.4(1)\times10^3$
	^{44}Ti	10(4)
	^{60}Co	<25
	^{137}Cs	$0.74(8)\times10^3$
	^{132}Cs	<15
	^{134}Cs	79(8)
	^{181}Hf	<11
	^{190}Pt	<20
	^{202}Pb	<9.1
^{232}Th	^{228}Ra	<12
	^{228}Th	<3.6
^{238}U	^{226}Ra	<23
	^{234}Th	<0.80
	^{234m}Pa	<0.48
^{235}U	^{235}U	<14

Radioactive contaminations of the CHC crystal measured with the ultra-low background **HP-Ge** γ spectrometer GeCris of the **STELLA** facility at LNGS.

The spectra of β/γ and α events selected by PSD analysis.



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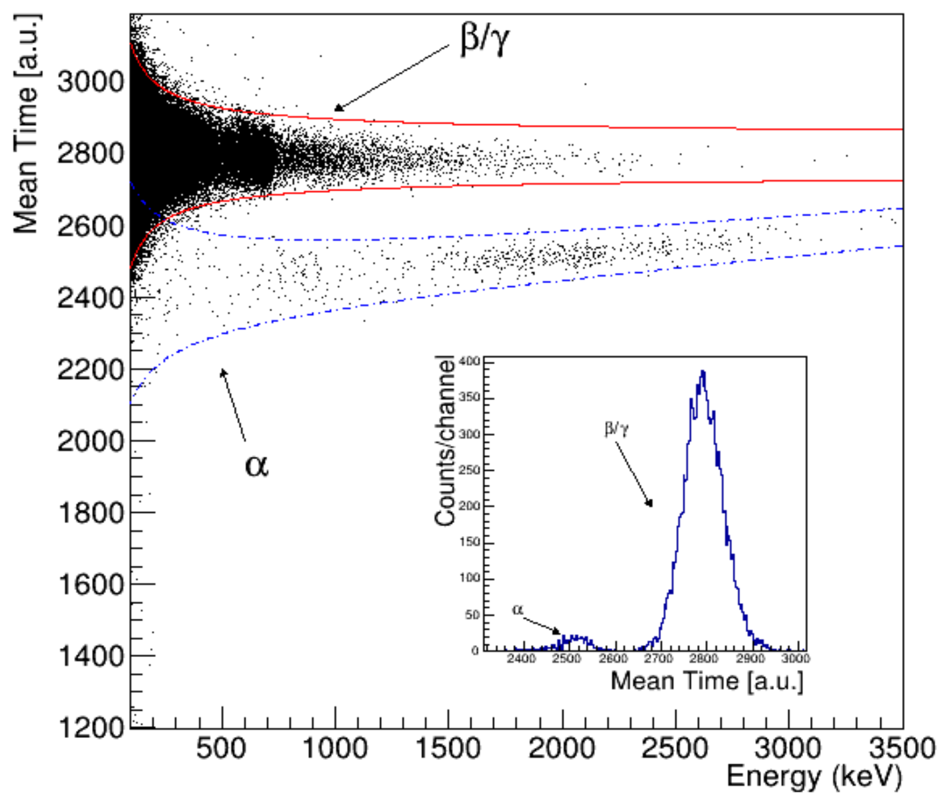
Data analysis

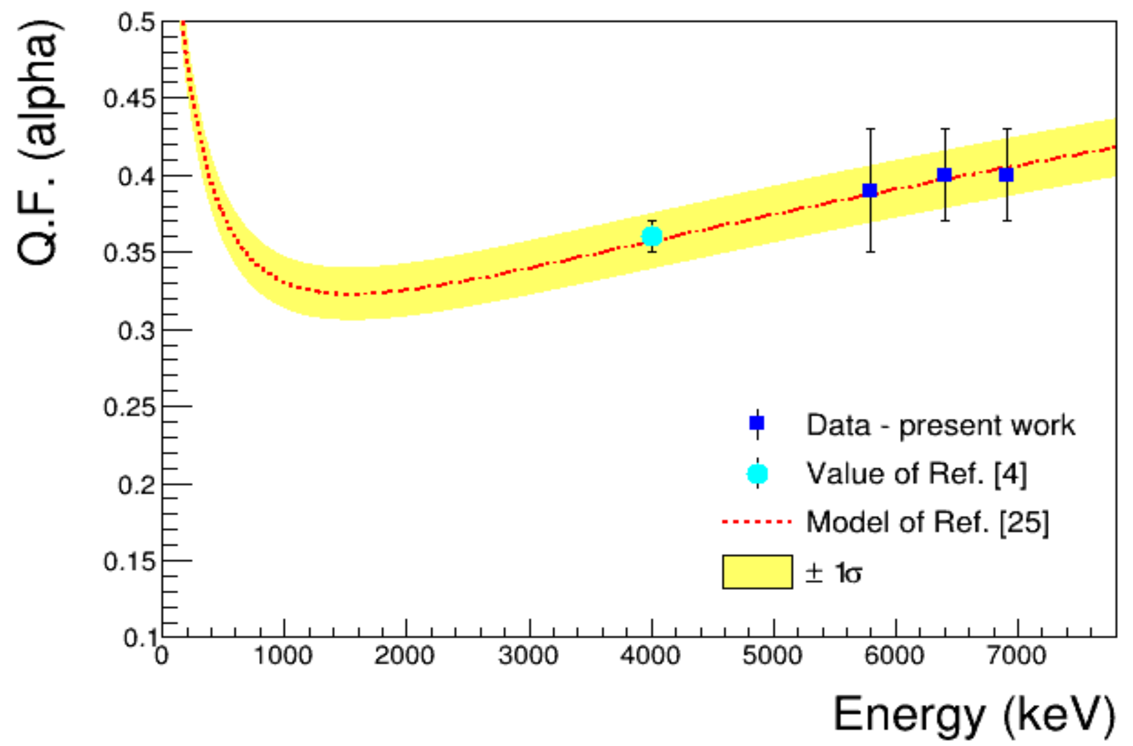
Time-amplitude analysis of ^{228}Th sub-chain and the derived Q.F.

[4] C. Cardenas, et al., Nucl. Instrum. Methods A 869 (2017) 63.
[25] V.I. Tretyak, Astropart. Phys. 33 (2010) 40.

Pulse Shape Discrimination (PSD) based on the pulse mean-time



The time-amplitude analysis was used to select the events of the following decay sub-chain of the ^{232}**Th family**:
^{224}**Ra** (Q = 5789 keV; $T_{1/2}$ = 3.66 d) → ^{220}**Rn** (Q = 6405 keV; $T_{1/2}$ = 55.6 s) → ^{216}**Po** (Q = 6906 keV; $T_{1/2}$ = 0.145 s) → ^{212}**Pb**.



The energies of the peaks of ^{224}**Ra**, ^{220}**Rn** and ^{216}**Po**, selected by the described time-amplitude analysis, are **2260(200) keV**, **2540(200) keV**, **2780(240) keV** (γ scale), respectively.

Results on the decay of naturally occurring Hf isotopes

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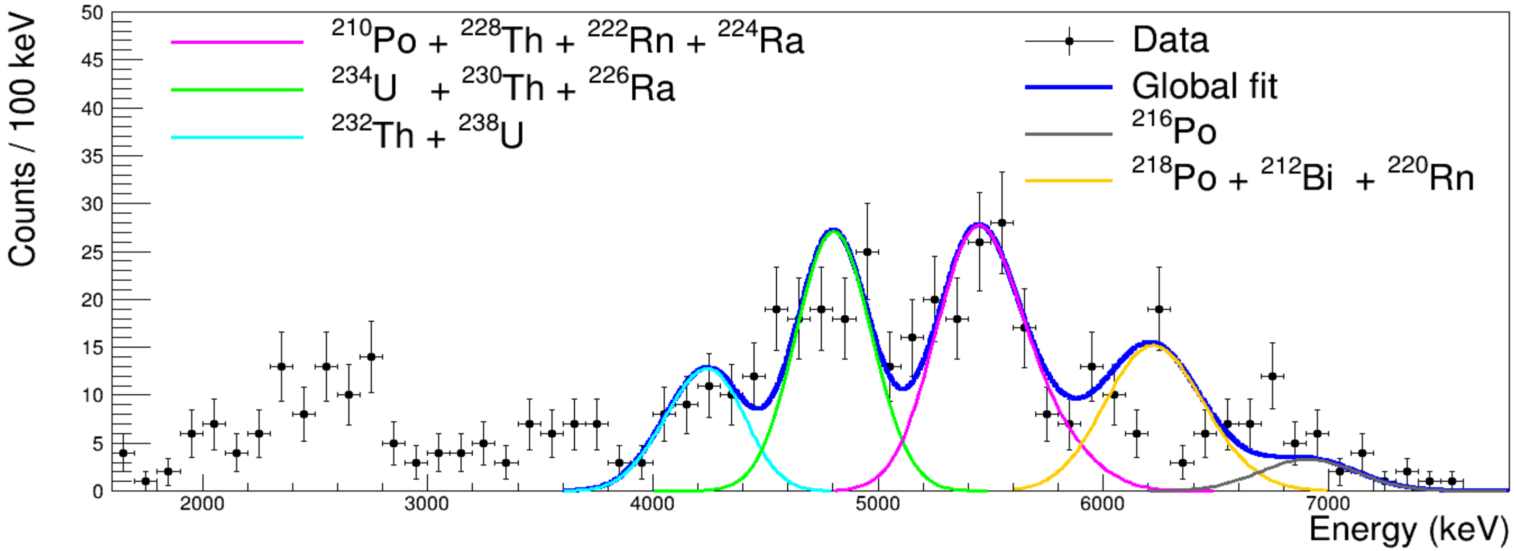
Energy spectrum of the α events selected by PSD from the data of the low-background measurements with the CHC crystal scintillator over 2848 h.



Considering all α events, the total internal α activity in the CHC crystal is at the level of **7.8(3) mBq/kg.**

Chain	Sub-Chain	Activity (mBq/kg)
^{232}Th	^{232}Th	0.2(1)
	^{228}Th	0.2(1)
^{238}U	^{238}U	0.6(1)
	^{234}U + ^{230}Th	1.4(2)
	^{226}Ra	0.2(1)
	^{210}Po	1.4(2)

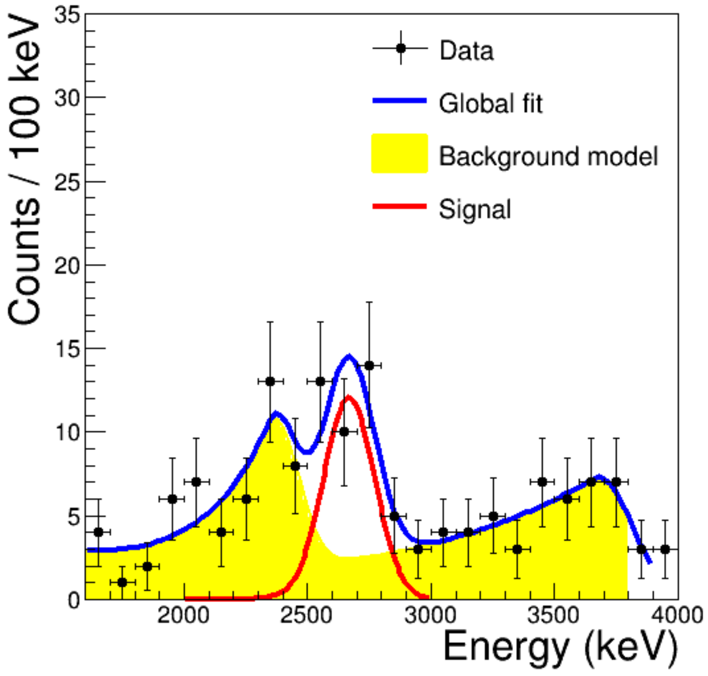
- In the hypothesis of the **half-life** of **1961** (T.P. Kohman, Phys. Rev. 121, 1758) **2.0(4)x10^{15} y** for the ^{174}Hf decay, the expected number of events (**2848** h of data taking) is about **1100** counts.
- The measured events are **553(23)** in total

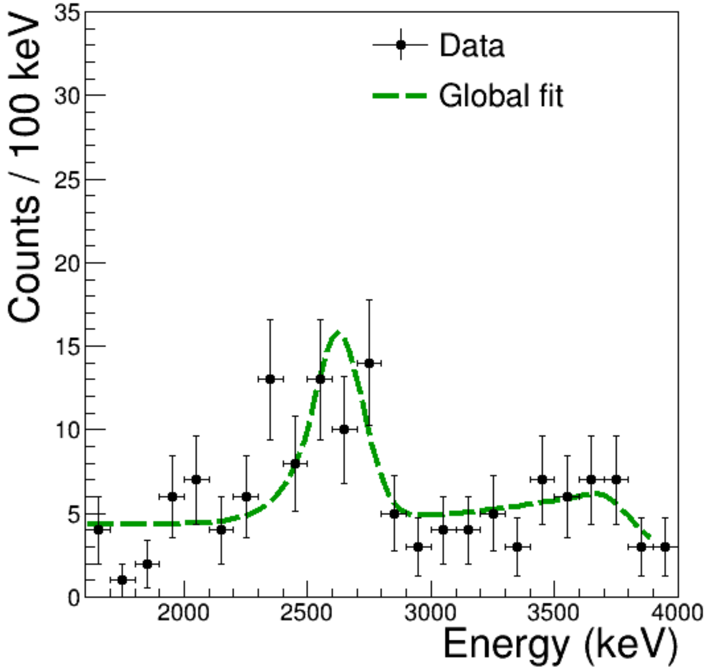
even ascribing all of them to ^{174}Hf decay, one **can safely rule out the old result for the T$_{1/2}$**

Results on the decay of naturally occurring Hf isotopes

χ^2/n.d.f.=0.87
P-value = 38.7%

Running-test,
tail probabilities:
Upper 94%
Lower 12%





χ^2 probability:
1.7%

The **background model** in the energy interval (1.1 – 3.9) MeV is made by an exponential function (to describe residual β/γ events), and suitable asym-Gaussian functions to describe the α decay of **^{147}Sm (Q_α = 2311.2(10) keV), ^{174}Hf (Q_α = 2494.5(2.3) keV)** and the events in the energy range (3.0-3.9) MeV. These events have been assumed to be degraded α events from possible surface and other contamination.

The counts for the peak near 2.3 MeV are **29.5(5.4)** in very good agreement with that expected for ^{147}Sm.

Half-lives on the α decay of Hf isotopes

(All the limits are given at 90% C.L.)

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Nuclide transition	Parent, daughter nuclei and its energy level (keV)	$T_{1/2}$ (y) Experimental: present work	previous works [10]	Theoretical: [7]	[8]	[9]
$^{174}Hf \to {}^{170}Yb$	$0^+ \to 0^+$, g.s.	$7.0 \pm 1.2 \times 10^{16}$	$2.0 \pm 0.4 \times 10^{15}$ [6,13]	$3.5 \cdot 10^{16}$	7.4×10^{16}	3.5×10^{16}
	$0^+ \to 2^+$, 84.3	$\geqslant 1.1 \times 10^{15}$	$\geqslant 3.3 \times 10^{15}$	$1.3 \cdot 10^{18}$	3.0×10^{18}	6.6×10^{17}
$^{176}Hf \to {}^{172}Yb$	$0^+ \to 0^+$, g.s.	$\geqslant 9.3 \times 10^{19}$	–	2.5×10^{20}	6.6×10^{20}	2.0×10^{20}
	$0^+ \to 2^+$, 78.7	$\geqslant 1.8 \times 10^{16}$	$\geqslant 3.0 \times 10^{17}$	1.3×10^{22}	3.5×10^{22}	4.9×10^{21}
$^{177}Hf \to {}^{173}Yb$	$7/2^- \to 5/2^-$, g.s.	$\geqslant 3.2 \times 10^{20}$	–	4.5×10^{20}	5.2×10^{22}	4.4×10^{22}
	$7/2^- \to 7/2^-$, 78.6	$\geqslant 7.5 \times 10^{16}$	$\geqslant 1.3 \times 10^{18}$	9.1×10^{21}	1.2×10^{24}	3.6×10^{23}
$^{178}Hf \to {}^{174}Yb$	$0^+ \to 0^+$, g.s.	$\geqslant 5.8 \times 10^{19}$	–	3.4×10^{23}	1.1×10^{24}	2.2×10^{23}
	$0^+ \to 2^+$, 76.5	$\geqslant 6.9 \times 10^{16}$	$\geqslant 2.0 \times 10^{17}$	2.4×10^{25}	8.1×10^{25}	7.1×10^{24}
$^{179}Hf \to {}^{175}Yb$	$9/2^+ \to 7/2^+$, g.s.	$\geqslant 2.5 \times 10^{20}$	$\geqslant 2.2 \times 10^{18}$	4.5×10^{29}	4.0×10^{32}	4.7×10^{31}
	$9/2^+ \to 9/2^+$, 104.5	$\geqslant 5.5 \times 10^{17}$	$\geqslant 2.2 \times 10^{18}$	2.0×10^{32}	2.5×10^{35}	2.2×10^{34}
$^{180}Hf \to {}^{176}Yb$	$9/2^+ \to 7/2^+$, g.s.	–	–	6.4×10^{45}	5.7×10^{46}	9.2×10^{44}
	$9/2^+ \to 9/2^+$, 82.1	–	$\geqslant 1.0 \times 10^{18}$	4.0×10^{49}	4.1×10^{50}	2.1×10^{48}

[6] T.P. Kohman, Phys. Rev. 121, 1758 (1961)

[7] B. Buck, A.C. Merchant, S.M. Perez, J. Phys. G 17 (1991) 1223.

[8] D.N. Poenaru, M. Ivascu, J. Phys. 44 (1983) 791.

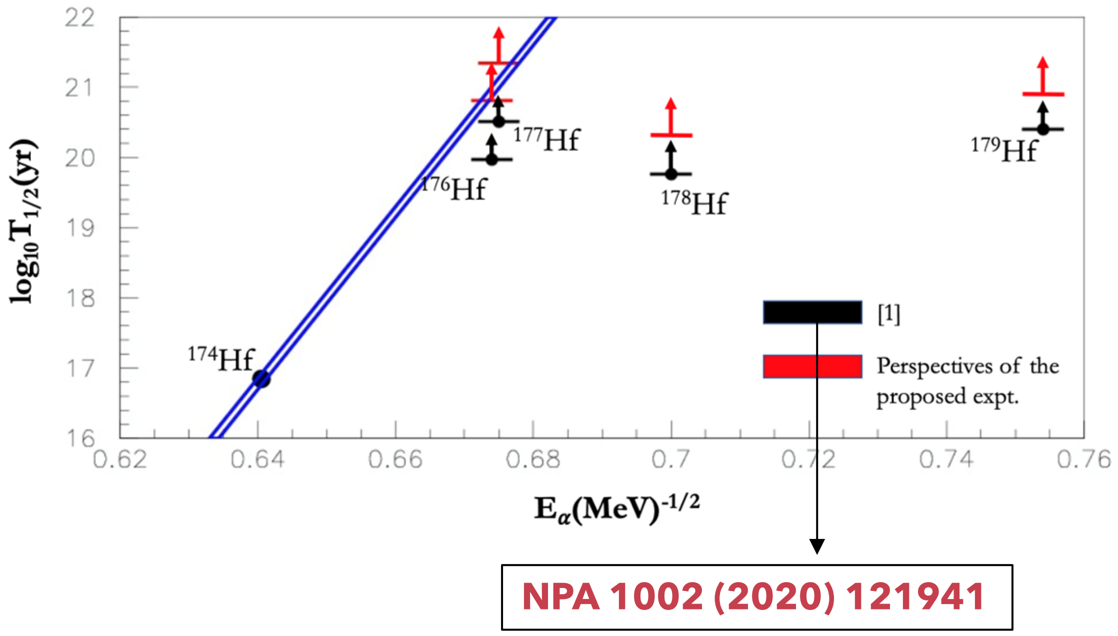
[9] V.Yu. Denisov, O.I. Davidovskaya, I.Yu. Sedykh, Phys. Rev. C 92 (2015) 014602.

[10] F. Danevich, et al., Eur. Phys. J. A 56 (5) (2020).

Perspectives and conclusions

- An experiment to measure low-level radioactive contaminants of a CHC crystal scintillator has been carried out at the STELLA facility at the LNGS.
- The results on rare nuclear transitions in Hf isotopes have led to rule out the $T_{1/2}$ value of the α decay of ^{174}Hf given in literature. In particular, we found that the α decay of ^{174}Hf to the ground state has been definitely observed with a **$T_{1/2}$ = 7.0(1.2) x 10^{16} y**. This value is in good agreement with the theoretical predictions.
- New lower limits of the half-life for **2ε** and **εβ⁺** decay of ^{174}Hf **(10^{16}-10^{18} y)** have been set.
- New lower limits of the half-life for *α* decay of ^{174}Hf to the first excited state and for *α* decays of ^{176}Hf, ^{177}Hf, ^{178}Hf, ^{179}Hf either to the ground states or to the first excited levels of daughter nuclides (**10^{16}-10^{20} y**) have been set.
- **Four CHC detectors, already at hand, will be fully characterized in the incoming months.**
- The expected results after 1 year of data taking of the four detectors will allow higher accuracy for the half-life of ^{174}Hf, α decay, of the order of 2.5%. Moreover, the sensitivity for the discovery of the α decay of the ^{176}Hf, ^{177}Hf isotopes will reach $T_{1/2}$~6.5×10^{20} yr and 2.2×10^{21} yr, well within the theoretical expectations reported in the previous table.
- In addition, these measurements will also be able to probe the α decay of the other Hf isotopes (^{179}Hf, ^{180}Hf).

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. **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. As evident, there is a good perspective to observe the *α* decay of ^{176}Hf and ^{177}Hf.