



Rare nuclear processes in Hf isotopes

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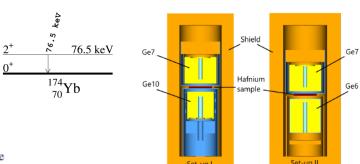
INTERESTING IN STUDYING THE 34 DBD+ EMITTERS

- DBD modes without the presence of neutrinos, if observed, can open windows on new physics
- to test calculations of different nucleus shapes and decay modes that involve the vector and axial-vector weak effective coupling constants; possible study of the "resonant effect" on the 0ν2ε mode;
- mutual information from the simultaneous study of positive and negative DBD can constrain the theoretical parameters with very high confidence
- the nuclear matrix elements for the two-neutrino mode and for the neutrinoless 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 ≈ 1.269 A^{-0.18} or g_A ≈ 1.269 A^{-0.12}, depending on the nuclear model adopted to infer the g_A value. DBD investigation with various nuclei would shed new light in constraining these and other important model-dependent parameters.
- As byproduct: developments of new detectors, e.g., new crystal scintillators containing DBD emitters

A search for double beta decays in hafnium using HP-Ge gamma spectrometer

 $\begin{array}{c|c}
\hline
 & 174 \\
 & 71 \\
\hline
 & 2\varepsilon, \varepsilon\beta^{+}
\end{array}$ $\begin{array}{c|c}
 & 0^{+} \\
 & 174 \\
 & 72 \\
\hline
 & P \\
 & Q_{2\beta} = 1100.0(23) \text{ keV}
\end{array}$

The isotope 174 Hf is one of the potentially 2ϵ , $\epsilon\beta^+$ radioactive nuclides with the energy of decay $Q_{2\beta}$ =1100.0(23)keV and the isotopic abundance δ =0.156(6)% (they used the old value 0.16(12)%).



The half-life limits on 2ε and $\varepsilon\beta^+$ processes in 174 Hf. The energies of the γ quanta (E_{γ}) , which were used to set the $T_{1/2}$ limits, are listed with their corresponding detection efficiencies (η) and values of $\lim S$.

 $m_{Hf} = 179.8 \text{ g}$, $\varnothing = 59.0 \text{ mm}$, h=5.0mm 225 m underground: Joint Research Centre of European Commission (Geel, Belgium). 75 days.

Channel of decay	Decay mode	Level of daughter nucleus (keV)	$\frac{E_{\gamma}}{(\text{keV})}$	η (%)	lim S (counts) at 90% C.L.	Experimental limit $T_{1/2}$ (a) at 90% C.L.
2 <i>K</i>	2ν	g.s.	51.4-61.0	1.24	36	$\geq 7.1 \times 10^{16}$
KL	2ν	g.s.	51.4-61.0	0.73	36	$\geq 4.2 \times 10^{16}$
2 <i>K</i>	2ν	2+ 76.5	51.4-61.0	1.03	36	$\geq 5.9 \times 10^{16}$
KL	2ν	2+ 76.5	51.4-61.0	0.61	36	$\geq 3.5 \times 10^{16}$
2L	2ν	2+ 76.5	76.5	0.39	20.4	$\geq 3.9 \times 10^{16}$
2 <i>K</i>	0ν	g.s.	977.4	4.53	10.0	$\geq 5.8 \times 10^{17}$
KL	0ν	g.s.	1028.9	4.46	3.0	$\geq 1.9 \times 10^{18}$
2L	0ν	g.s.	1080.4	4.39	7.2	$\geq 7.8 \times 10^{17}$
2 <i>K</i>	0ν	2+ 76.5	900.9	4.67	8.4	$\geq 7.1 \times 10^{17}$
KL	0ν	2+ 76.5	952.4	4.59	9.5	$\geq 6.2 \times 10^{17}$
2L	0ν	0+ 76.5	1003.9	4.51	8.0	$\geq 7.2 \times 10^{17}$
$K\beta^+$	(2v + 0v)	g.s.	511	10.6	202	$\geq 1.4 \times 10^{17}$
$L\beta^+$	(2v + 0v)	g.s.	511	10.7	202	$\geq 1.4 \times 10^{17}$

Chain	Nuclide	Activity in the sample (mBq)
	⁴⁰ K	≤ 1.4
	⁶⁰ Co	≤ 0.11
	¹³⁷ Cs	≤ 0.20
	¹⁷² Hf	≤3
	¹⁷⁵ Hf	$0.44 \pm 0.05 \ (0.18 \pm 0.04)$
	178m2Hf	≤ 0.06
	¹⁸¹ Hf	$1.45 \pm 0.07 \ (0.12 \pm 0.04)$
	¹⁸² Hf	≤ 0.5
²³² Th	²²⁸ Ra	3.6 ± 0.7
	²²⁸ Th	2.38 ± 0.25
235U	²³⁵ U	3.8 ± 0.5
	231 Pa	11 ± 3
	²²⁷ Ac	2.0 ± 0.5
²³⁸ U	234m Pa	11 ± 5
	²²⁶ Ra	≤ 0.7
	²¹⁰ Pb	_ ≤ 50

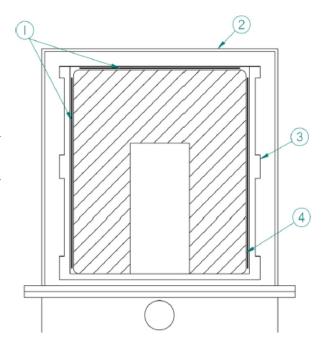
Nuclear Physics A 996 (2020) 121703

In case of the 2K and KL capture in 174 Hf, a cascade of X-rays (and Auger electrons) of Yb atom with individual energies, in particular, in the energy interval (50.8–61.3)keV is expected, while energies of the 2L capture X-ray quanta are \approx (7 –10)keV, that are below the detectors' energy thresholds. We took into account only the most intense X-rays of ytterbium: 51.4 keV (the intensity of the X-ray quanta is 27.2%), 52.4 keV (47.4%), 59.2 keV (5.2%), 59.4 keV (10.0%), and 61.0 keV (3.4%).

A search for double beta decays in hafnium using HP-Ge gamma spectrometer

Decay Mode	$E_{\gamma,X}$ [keV]	η [%]	Experimental $T_{1/2}$ [a] This work
2v2K (g.s.)	52.4	0.50	$\geq 1.4 \times 10^{16}$
$2\nu KL$ (g.s.)	52.4	0.50	$\geq 1.4 \times 10^{16}$
$2\nu 2K$ (1st exc.)	76.5	3.15	$\geq 7.9 \times 10^{16}$
$2\nu 2K$ (1st exc.)	52.4	0.50	$\geq 1.4 \times 10^{16}$
$2\nu KL$ (1st exc.)	76.5	3.15	$\geq 7.9 \times 10^{16}$
$2\nu KL$ (1st exc.)	52.4	0.50	$\geq 1.4 \times 10^{16}$
$2\nu 2L$ (1st exc.)	76.5	3.15	$\geq 7.9 \times 10^{16}$
0v2K (g.s.)	977.4	7.59	$\geq 2.7 \times 10^{18}$
0v2K (1st exc.)	900.9	8.01	$\geq 2.4 \times 10^{18}$
$0\nu KL$ (g.s.)	1028.9	7.32	$\geq 4.2 \times 10^{17}$
$0\nu KL$ (1st exc.)	952.4	7.72	$\geq 3.1 \times 10^{17}$
$0\nu LL$ (g.s.)	1080.4	7.09	$\geq 3.6 \times 10^{17}$
$0\nu LL$ (1st exc.)	1003.9	7.45	$\geq 9.4 \times 10^{17}$
$K\beta^+ (0\nu + \nu\bar{\nu})$	511	11.8	$\geq 5.6 \times 10^{16}$
$L\beta^+ (0\nu + \nu\bar{\nu})$	511	11.8	$\geq 5.6 \times 10^{16}$

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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.

Hf foil: 0.25(1) mm thick, m=55,379(1) g, 310 days.

INTERESTING IN STUDYING RARE & 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 halflives.
- The study on the nuclear instability offers information 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 of α particles through the barrier, caused by the interaction between the α and the nucleus
- Among the naturally occurring α -emitting nuclides only those with either A > 208 or A \approx 145 have α half-lives short enough to be detected
- As byproduct: developments of new detectors, e.g., new crystal scintillators containing α emitters

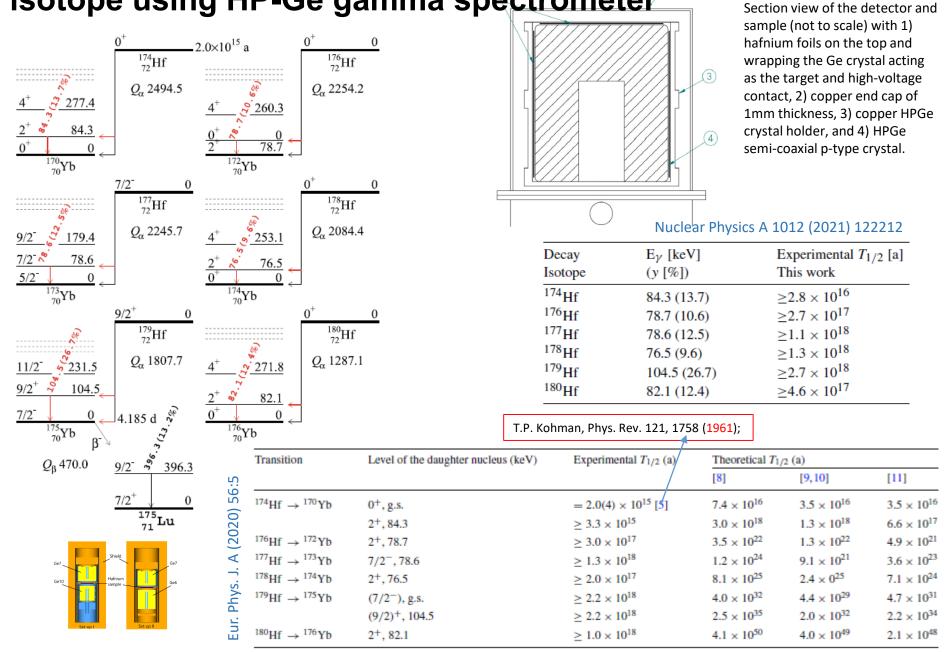
Potentially α decay of naturally occurring Hf-nuclides

Some potential transitions of Hf isotopes and related information. Only naturally occurring isotopes (with natural abundance δ) and with Q > 0 between g.s. transitions or between g.s. and lowest bound level transitions (with spin/parity J^{π}) are listed. E is the kinetic energy of the alpha particle. N is the number of nuclei in the CHC crystal used in this work. Experimental measurements (when available) and theoretical prediction of the half-live are reported in the last four columns.

Nuclide Transition	J^{π} of	δ	Q_{α}	E_{lpha}	N		$T_{1/2}$ (y)	
	Parent \rightarrow Daughter Nuclei and its level (keV) [10, 11]	(%) [2]		(keV)		Experimental	[15]	Theoretical [16]	[9]
$^{174}\mathrm{Hf} ightarrow ^{170}\mathrm{Yb}$	$0^+ \to 0^+$, g.s. $0^+ \to 2^+$, 84.2	0.16(12)	2494.5(2.3)	2437.6(2.2)	1.0×10^{19}	$2.0(4) \times 10^{15} [6.13]$ $\geqslant 3.3 \cdot 10^{15} [14]$	$3.5 \cdot 10^{16}$ $1.3 \cdot 10^{16}$	7.4×10^{16} 3.0×10^{18}	3.5×10^{16} 6.6×10^{17}
$^{176}\mathrm{Hf} ightarrow ^{172}\mathrm{Yb}$	$0^{+} \rightarrow 0^{+}, \text{ g.s.}$ $0^{+} \rightarrow 2^{+}, 78.7$	5.26(70)	2254.2(1.5)	2203.3(1.5)	3.3×10^{20}	$> 3.0 \times 10^{17} [14]$	2.5×10^{20} 1.3×10^{22}		2.0×10^{20} 4.9×10^{21}
$^{177}\mathrm{Hf} ightarrow ^{173}\mathrm{Yb}$	$7/2^- \rightarrow 5/2^-$, g.s. $7/2^- \rightarrow 7/2^-$, 78.6	18.60(16)	2245.7(1.4)	2195.3(1.4)	1.2×10^{21}	$\geqslant 1.3 \times 10^{18} \ [14]$	$\begin{array}{c} 4.5 \times 10^{20} \\ 9.1 \times 10^{21} \end{array}$	$5.2 \times 10^{22} \\ 1.2 \times 10^{24}$	$4.4 \times 10^{22} \\ 3.6 \times 10^{23}$
$^{178}\mathrm{Hf} ightarrow ^{174}\mathrm{Yb}$	$0^+ \to 0^+$, g.s. $0^+ \to 2^+$, 76.5	27.28(28)	2084.4(1.4)	2037.9(1.4)	1.7×10^{21}	$\geqslant 2.0 \times 10^{17} \ [14]$	3.4×10^{23} 2.4×10^{25}	$1.1 \times 10^{24} \\ 8.1 \times 10^{25}$	$2.2 \times 10^{23} \\ 7.1 \times 10^{24}$
$^{179}\mathrm{Hf} ightarrow ^{175}\mathrm{Yb}$	$9/2^+ \rightarrow 7/2^+$, g.s. $9/2^+ \rightarrow 9/2^+$, 104.5	13.62(11)	1807.7(1.4)	1767.6(1.4)	8.6×10^{20}	$\geqslant 2.2 \times 10^{18} $ [14] $\geqslant 2.2 \times 10^{18} $ [14]	$4.5 \times 10^{29} \\ 2.0 \times 10^{32}$	$4.0 \times 10^{32} \\ 2.5 \times 10^{35}$	$4.7 \times 10^{31} \\ 2.2 \times 10^{34}$
$^{180}\mathrm{Hf} ightarrow ^{176}\mathrm{Yb}$	$0^+ \to 0^+, \text{ g.s.}$ $0^+ \to 2^+, 82.1$	35.08(33)	1287.1(1.4)	1258.7(1.4)	2.2×10^{21}	$\geqslant 1.0 \times 10^{18} \boxed{14}$	$6.4 \times 10^{45} 4.0 \times 10^{49}$	$5.7 \times 10^{46} 4.1 \times 10^{50}$	9.2×10^{44} 2.1×10^{48}
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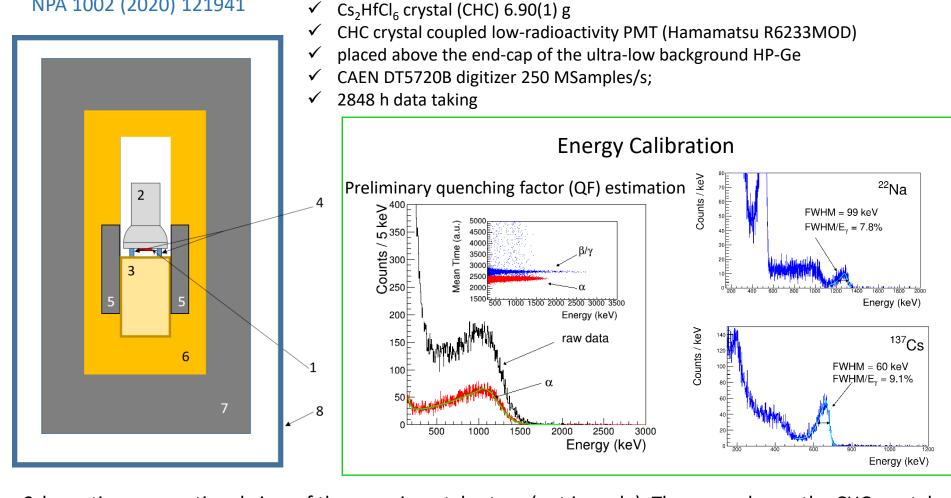
T.P. Kohman, Phys. Rev. 121, 1758 (1961);

A search for alpha decays in excited states of hafnium isotope using HP-Ge gamma spectrometer section view of the



Study of rare alpha decay in Hf isotopes using a crystal scintillators (source=detector): the experiment

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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 (5), high purity copper (6), low radioactive lead (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-N₂ gas.

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

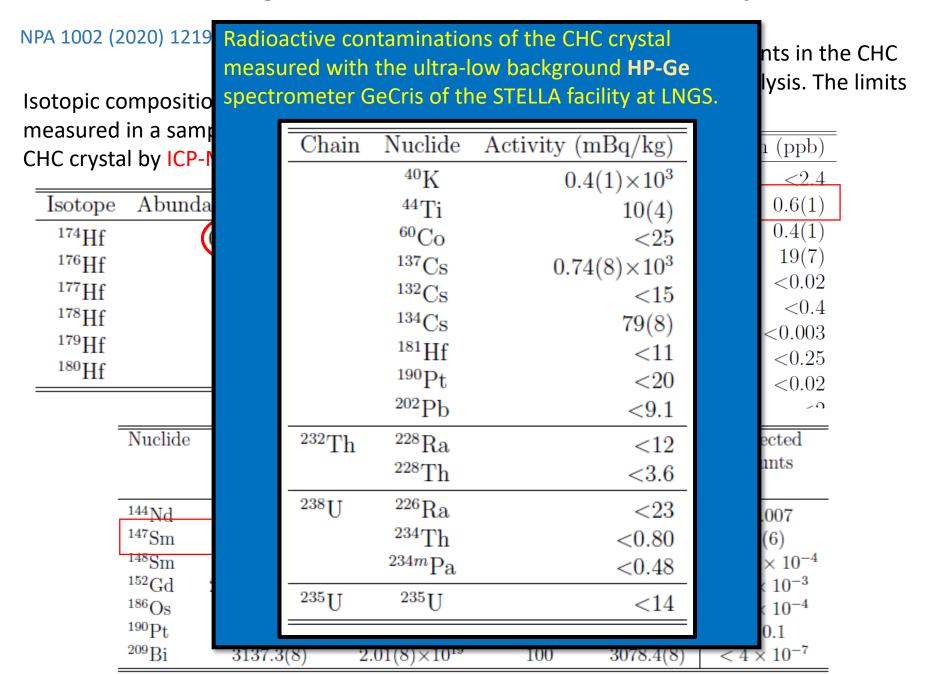
Isotope	Abundance (%)
¹⁷⁴ Hf	(0.156(6))
$^{176}\mathrm{Hf}$	5.18(5)
$^{177}\mathrm{Hf}$	18.5(1)
$^{178}\mathrm{Hf}$	27.2(1)
$^{179}{ m Hf}$	13.9(1)
¹⁸⁰ Hf	35.2(2)

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

:	Nuclide	Concentration (ppb)
	$^{144}\mathrm{Nd}$	< 2.4
	$^{147}\mathrm{Sm}$	0.6(1)
	$^{148}\mathrm{Sm}$	0.4(1)
	$^{151}\mathrm{Eu}$	19(7)
	$^{152}\mathrm{Gd}$	< 0.02
	$^{180}\mathrm{W}$	< 0.4
	$^{184}\mathrm{Os}$	< 0.003
	$^{186}\mathrm{Os}$	< 0.25
	$^{190}\mathrm{Pt}$	< 0.02
	209 Bi	<2

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

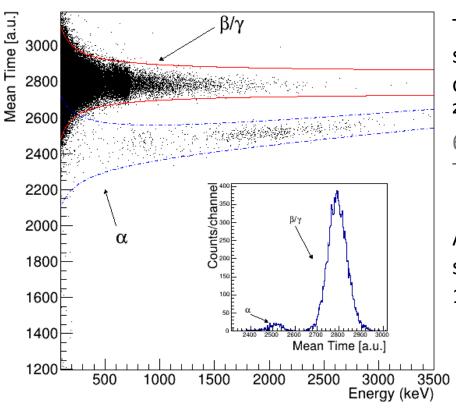
Low background measurements of the CHC crystal



Data analysis

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

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

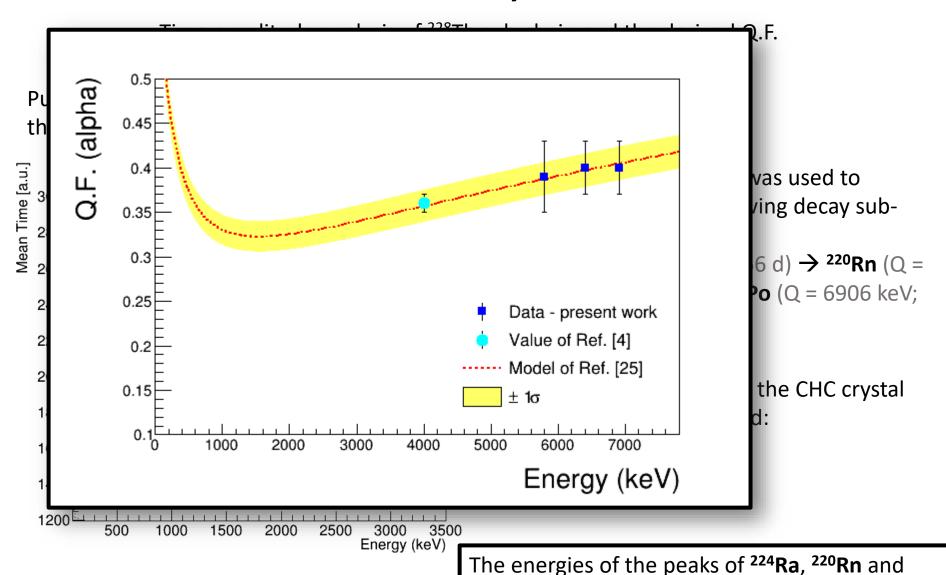


The time-amplitude analysis was used to select the events of the following decay subchain of the ²³²Th family:

²²⁴Ra (Q = 5789 keV; $T_{1/2}$ = 3.66 d) → ²²⁰Rn (Q = 6405 keV; $T_{1/2}$ = 55.6 s) → ²¹⁶Po (Q = 6906 keV; $T_{1/2}$ = 0.145 s) → ²¹²Pb.

An average activity of 228 Th in the CHC crystal scintillator has been estimated: $100(50) \mu Bq/kg$

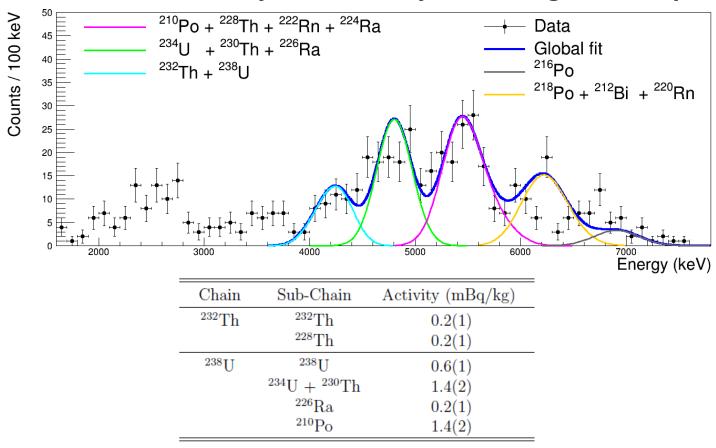
Data analysis



²¹⁶Po, selected by the described time-amplitude analysis, are 2260(200) keV, 2540(200) keV, 2780(240) keV (γ scale), respectively.

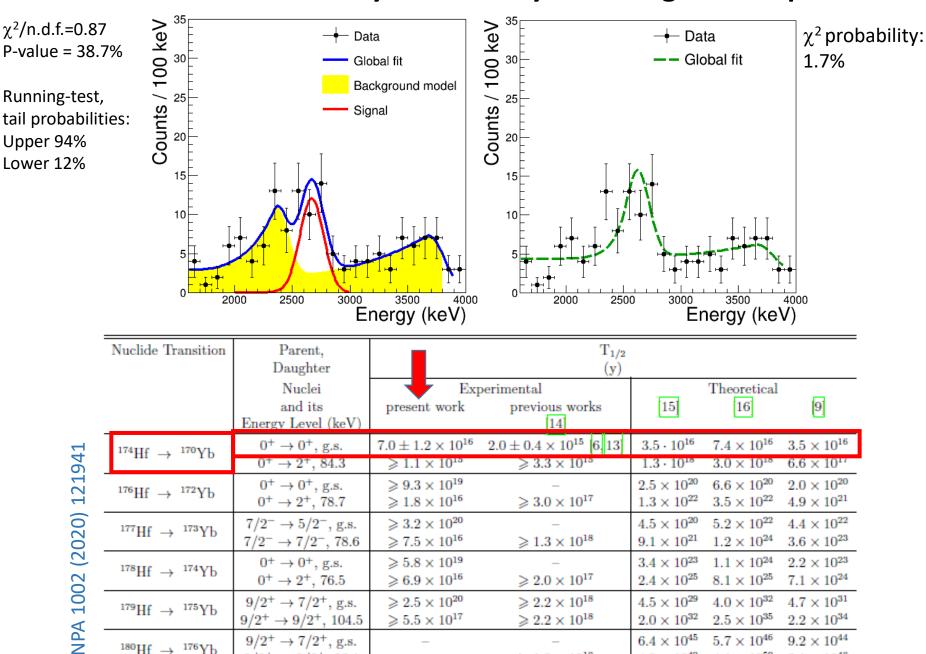
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Results on the decay of naturally occurring Hf isotopes



When adopting the claimed half-life 2.0(4) 10^{15} y for the 174 Hf decay, the expected number of events - within 2848 h of data taking with the used CHC crystal - is about 1100 counts. Thus, considering that the measured events are 553(23) in total, even ascribing all of them to 174 Hf decay (despite the analysis reported above), one can safely rule out the result of 2.0(4) 10^{15} y; in fact, even in such an unlike hypothesis, the $T_{1/2}$ value derived from the present experimental data would be 4.01(17) 10^{15} y, i.e. is about 4.5 σ far from the value 2.0(4) 10^{15} y. Thus, the $T_{1/2}$ value 2.0(4) 10^{15} y is safely rejected. Let us now perform a more refined determination of the $T_{1/2}$ value of the 174 Hf decay supported by our data.

Results on the decay of naturally occurring Hf isotopes



 $\geq 1.0 \times 10^{18}$

 4.0×10^{49}

 4.1×10^{50}

 2.1×10^{48}

 $9/2^+ \rightarrow 7/2^+$, g.s.

 $9/2^+ \rightarrow 9/2^+, 82.1$

 $^{180}\mathrm{Hf} \rightarrow ^{176}\mathrm{Yb}$

Conclusion

To study the decay of naturally occurring hafnium to the ground state and to the first excited states, a CHC crystal scintillator was used in coincidence with a HP-Ge detector in 2848 h of live time.

- The results rules out the $T_{1/2}$ value of the alpha decay of ¹⁷⁴Hf given in literature. In particular, we found that the decay of ¹⁷⁴Hf to the ground state has been definitely observed with a $T_{1/2} = 7.0(1.2) \times 10^{16} \text{ y}$. This value is in good agreement with the theoretical predictions.
- New lower limits of the half-life for alpha decay of ¹⁷⁴Hf to the first excited state and for alpha decays of ¹⁷⁶Hf, ¹⁷⁷Hf, ¹⁷⁸Hf, ¹⁷⁹Hf either to the ground states or to the first excited levels of daughter nuclides (10¹⁶-10²⁰ y) have been set.
- \triangleright New lower limits of the half-life for 2ε and εβ⁺ decay of ^{174}Hf ($10^{16}-10^{18}$ y) have been set.