Investigation of rare nuclear processes in neodymium and osmium naturally occurring isotopes

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The investigations has been done in collaboration with:

- DAMA group (Rome, Italy)
- National Science Center 'Kharkiv Institute of Physics and Technology' and V.N. Karazin Kharkiv National University (Kharkiv, Ukraine)
- Institute of Theoretical and Experimental Physics, National Research Centre 'Kurchatov Institute' (Moscow, Russia)
- John de Laeter Centre for Isotope Research, Curtin University (Bentley, Australia)

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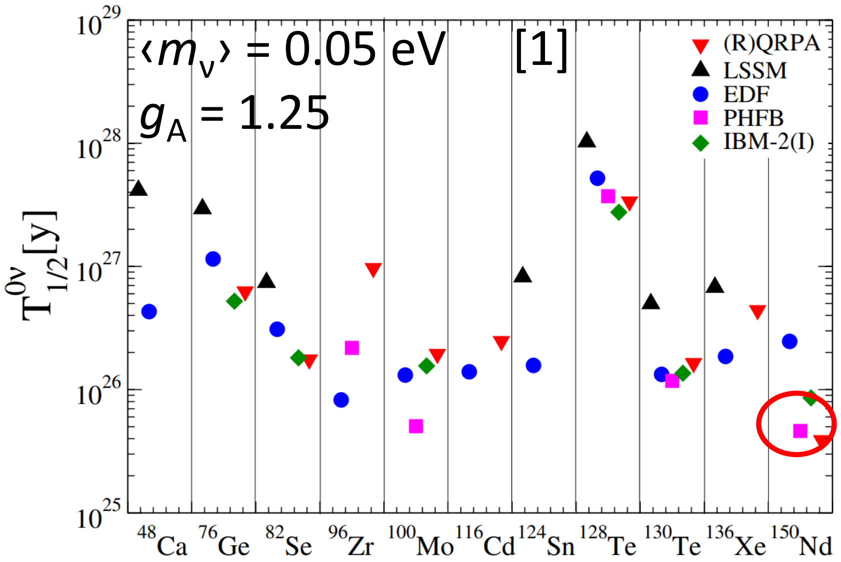
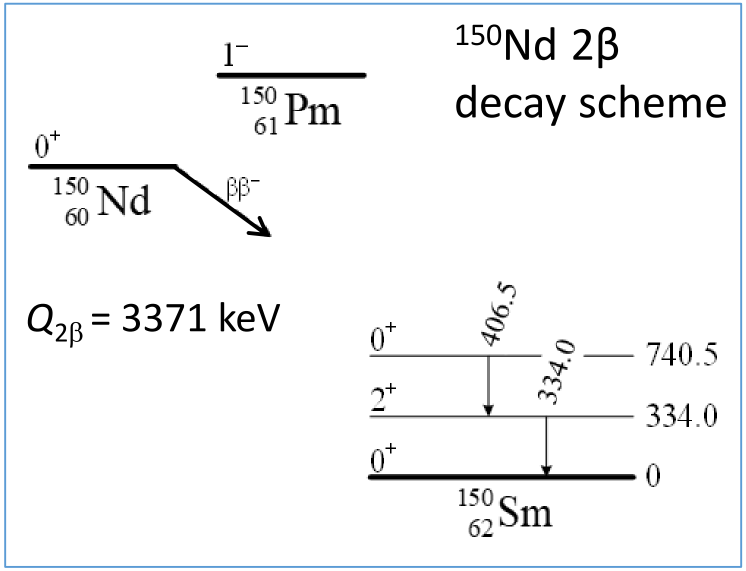
1. Investigation of double beta decay of ^{150}Nd to the first 0^+ excited level of ^{150}Sm (E^*=740.5 keV)

^{150}Nd: one of the most promising nuclides for 2β experiments





- High energy release

  $Q_{\beta\beta}$ = 3371.38(20) keV [2]

- Optimistic theoretical estimations of $T_{1/2}$
- Comparatively high natural isotopic abundance

  δ = 5.638(28)% [3]

- Possibility to investigate the decay to excited levels of ^{150}Sm

[1] J.D. Vergados et al., Rep. Prog. Phys. 75 (2012) 106301
[2] V.S. Kolhinen et al., Phys. Rev. C 82 (2010) 022501
[3] J. Meija et al., Pure Appl. Chem. 88 (2016) 293

Previous observations of ^{150}Nd → ^{150}Sm (0^+, 740.5 keV) transition

Short description	$T_{1/2}$, 10^{20} y #	Year
Modane underground laboratory (4800 m w.e.), HP Ge 400 cm^3, 3046 g of Nd_2O_3 (δ = 5.638%), 11321 h, 1-dim spectrum	$1.4^{+0.4}_{-0.2} \pm 0.3$	2004 [1]
Re-estimation of the result [1]	$\mathbf{1.33^{+0.36}_{-0.23}\,^{+0.27}_{-0.13}}$	**2009 [2]**
Modane underground laboratory (4800 m w.e.), NEMO-3 detector, foil with 57.2 g of $^{150}Nd_2O_3$ (δ = 91.0%), 40774 h, energies of e^- and γ, tracks for e^-	$0.71 \pm 0.13 \pm 0.09$	2013 [3]
Kimballton Underground Research Facility (1450 m w.e.), 2 HPGe (~304 cm^3 each one), 50 g $^{150}Nd_2O_3$ (δ = 93.6%), 15427 h, coincidence spectrum	$\mathbf{1.07^{+0.45}_{-0.25} \pm 0.07}$	**2014 [4]**
NEMO-3 (re-estimation of [3])	$1.11^{+0.19}_{-0.14}\,^{+0.17}_{-0.15}$	2021 [5]

The 1st uncertainty is statistical, the 2nd one corresponds to systematics

[1] A.S. Barabash et al., *Phys. Atom. Nucl.* **67** (2004) 1216.

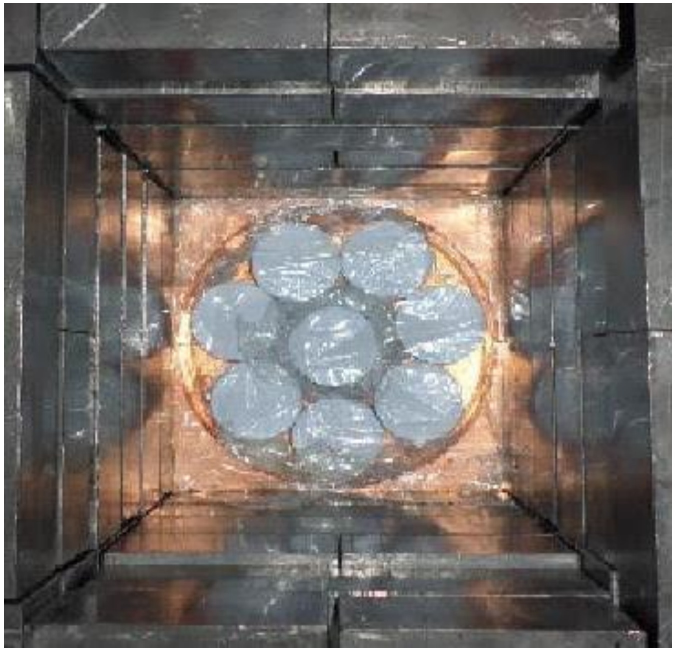
[2] A.S. Barabash et al., *Phys. Rev. C* **79** (2009) 045501.

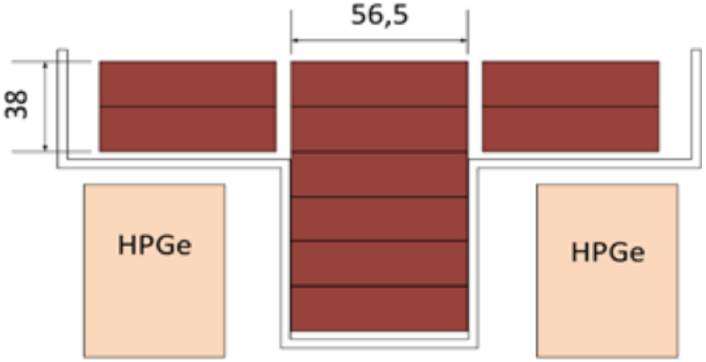
[3] S. Blondel, PhD thesis, LAL, Orsay, France (2013).

[4] M.F. Kidd et al., *Phys. Rev. C* **90** (2014) 055501.

[5] V. Tretyak, LXXI Int. Conf. "NUCLEUS-2021", 20-25 Sep 2021, Book of Abstracts, Saint Petersburg (2021), p. 257

Experimental setup



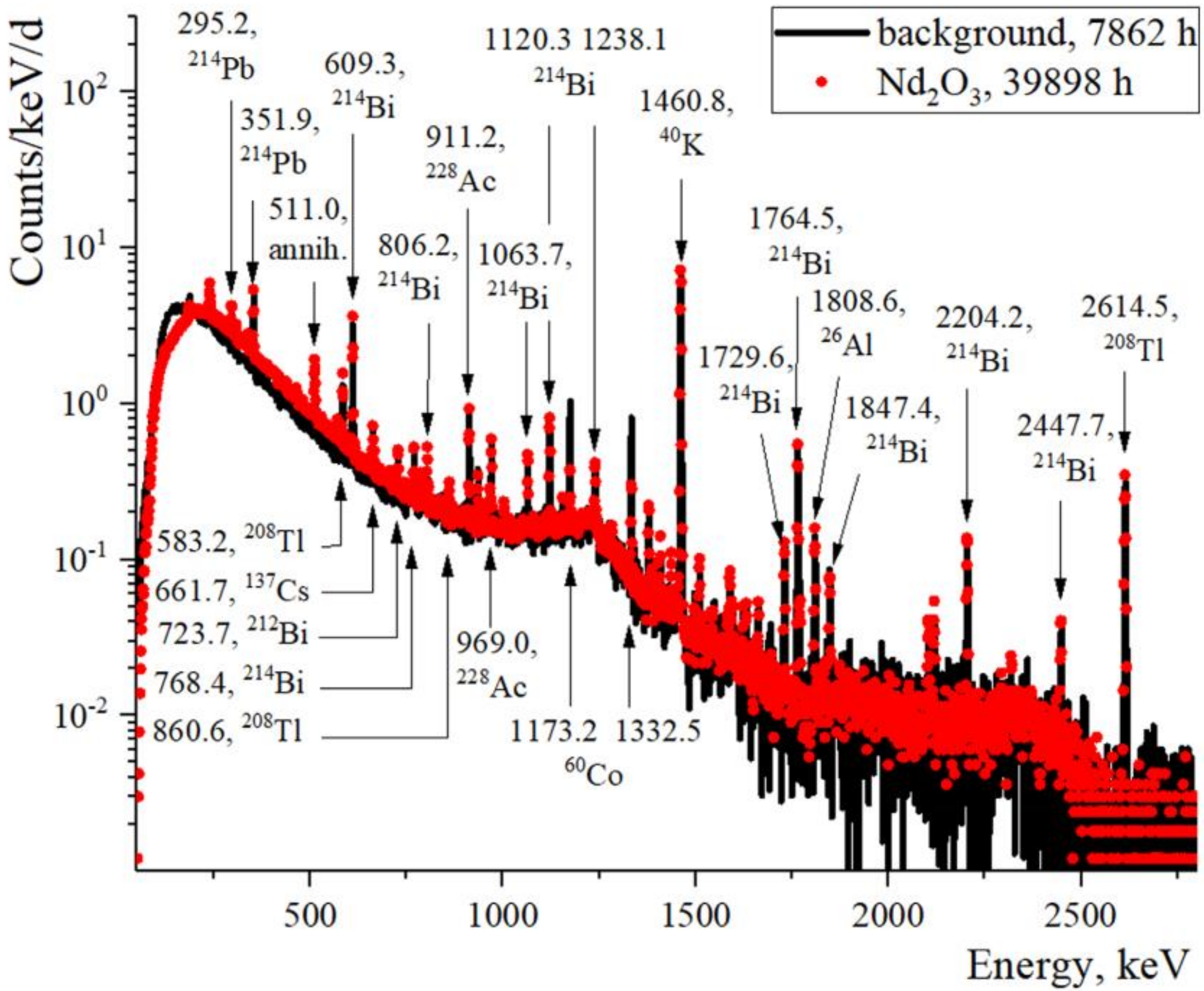


- 2381-g Nd_2O_3 sample (average density ~2.84 g/cm^3), used in previous experiment [1], additionally purified before the present measurements [2]
- 4 HP Ge detectors (≃225 cm^3 each) in a cryostat with cylindrical well in the center; Gran Sasso National Laboratory (LNGS)
- Shield: copper (10 cm), lead (20 cm)
- Plexiglas container flushed with high-purity nitrogen gas (to remove radon)

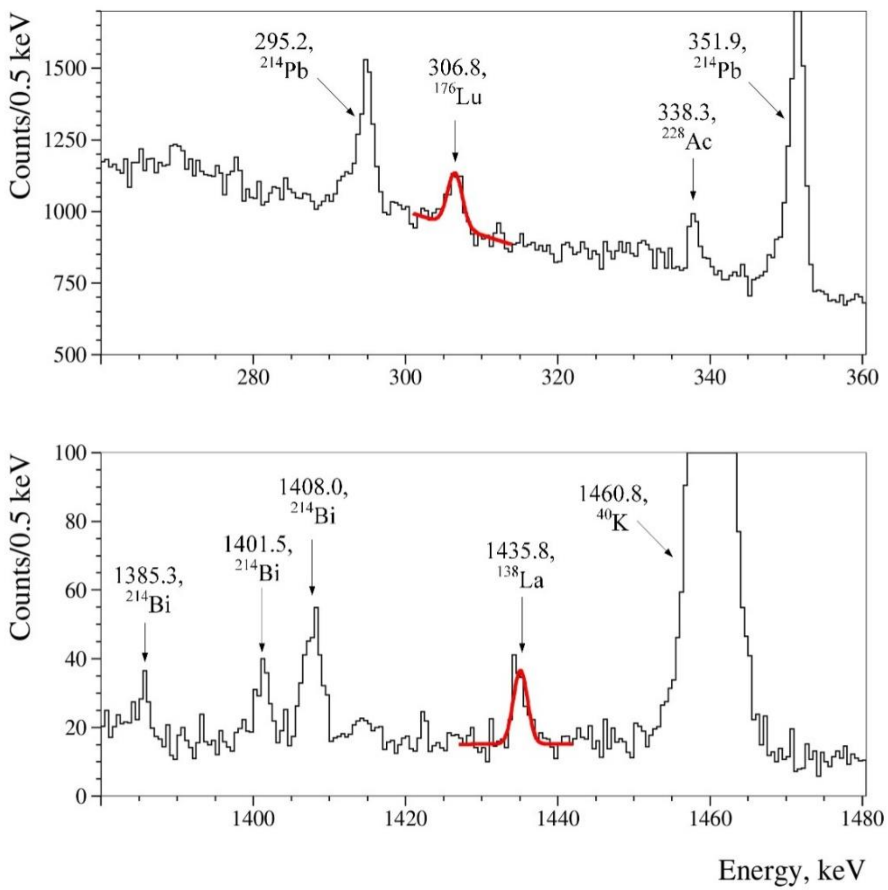
No. of detector	FWHM, keV (1333 keV, ^{60}Co calibration source)
1	2.36(2)
2	2.01(2)
3	2.06(2)
4	4.01(4)

[1] A.S. Barabash et al., Phys. Atom. Nucl. 67 (2004) 1216.
[2] R.S. Boiko, Int. J. Mod. Phys. A 32 (2017) 1743005.

Nd_2O_3 vs background

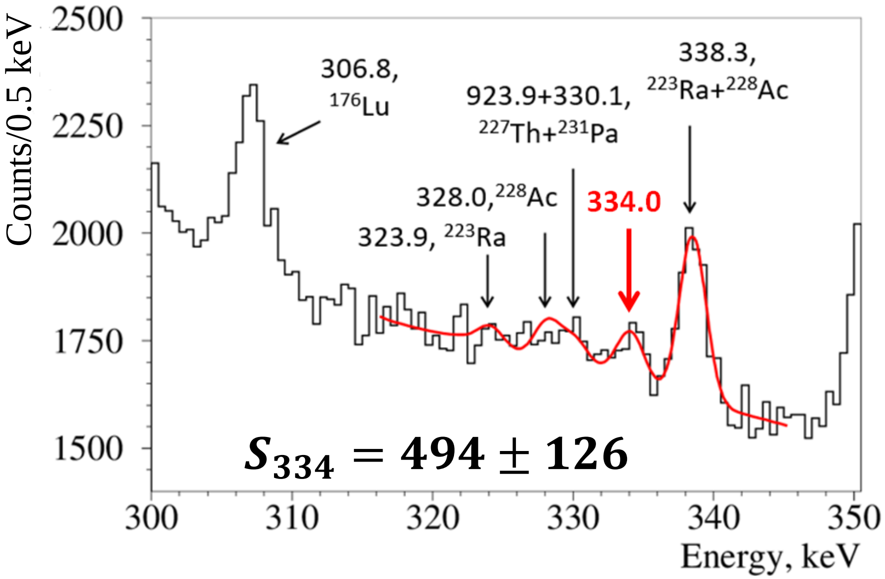


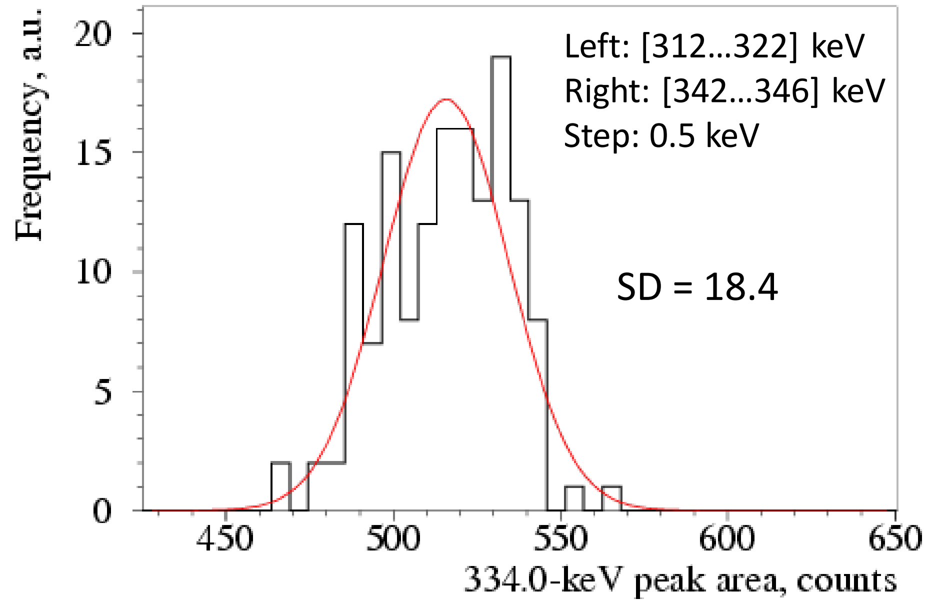
Radioactive contamination of the Nd_2O_3 sample



Chain	Nuclei	Activity, mBq/kg	
		Before purification	After purification
	^{40}K	16(8)	**≤ 1.8**
	^{137}Cs	≤ 0.8	≤ 0.04
	^{138}La	–	0.057(9)
	^{176}Lu	1.1(4)	**0.29(4)**
^{232}Th	^{228}Ra	≤ 2.1	≤ 0.3
	^{228}Th	≤ 1.3	≤ 0.4
^{235}U	^{235}U	≤ 1.7	≤ 1.3
^{238}U	^{234}Th	≤ 28	≤ 5.4
	^{226}Ra	1.5(8)	≤ 1.9

1-dim spectrum analysis (334.0 keV)



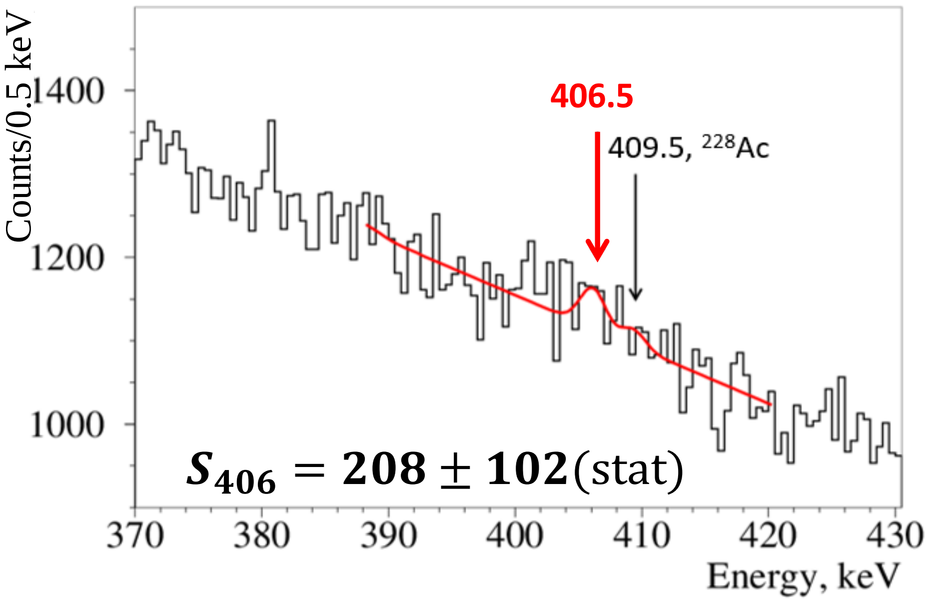


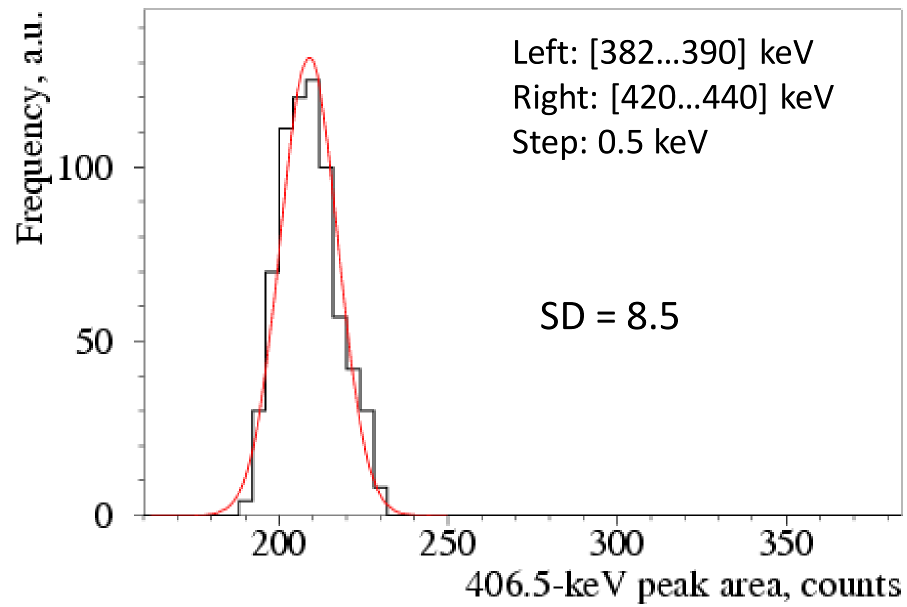
Frequency distribution of S_{334} with different fit intervals

Systematic	ΔS_{334}, counts
Interval of fit	± 18.4
MC uncertainty (10%)	± 49.4
Number of ^{150}Nd nuclei (0.5%)	± 2.5

$$T_{1/2}^{334} = [6.6^{+2.3}_{-1.4}(\text{stat}) \pm 0.8\ (\text{syst})] \cdot 10^{19}\text{y}$$

1-dim spectrum analysis (406.5 keV)



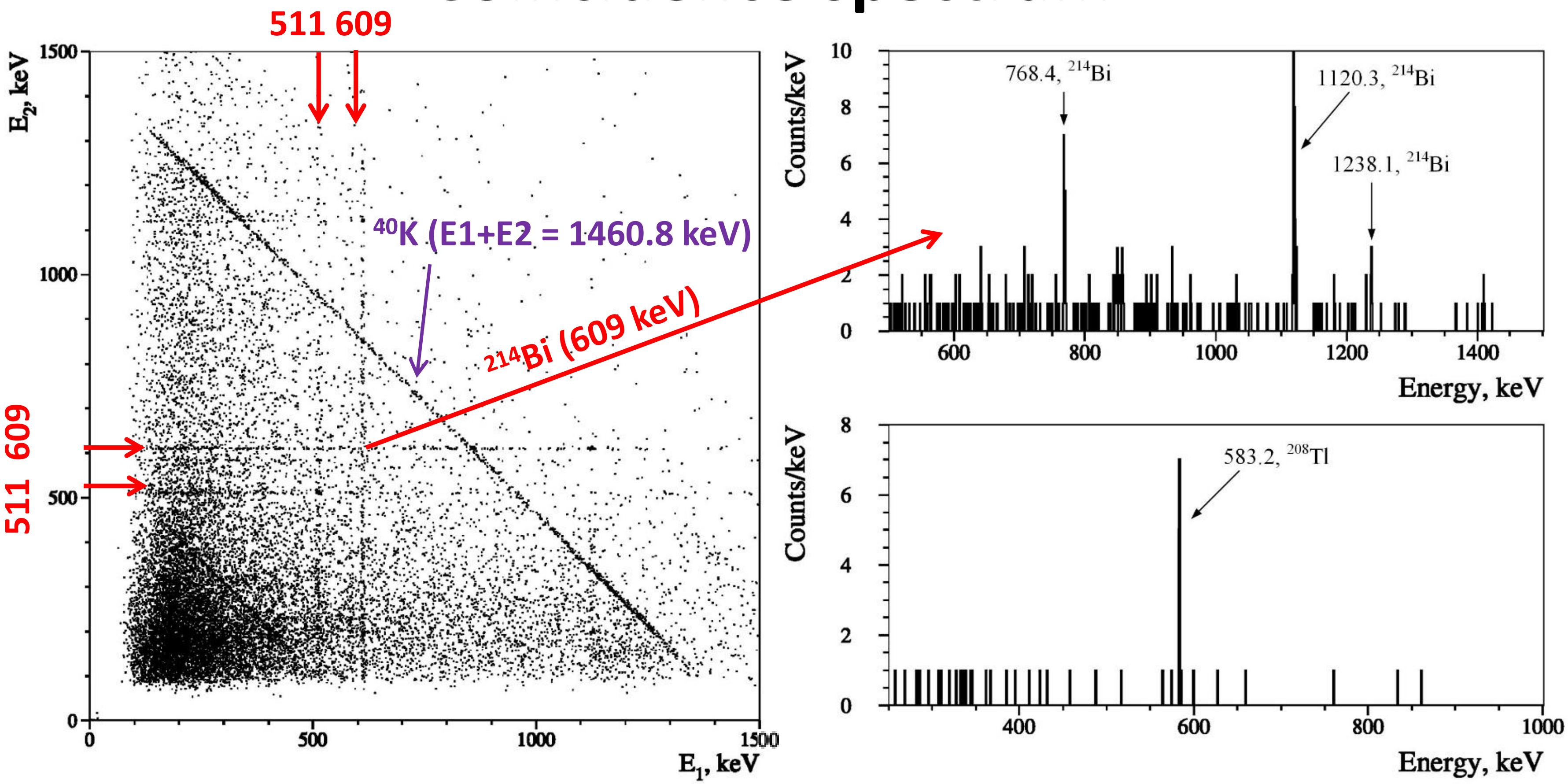


Frequency distribution of S_{406} with different fit intervals

Systematic	ΔS_{406}, counts
Interval of fit	± 8.5
MC uncertainty (10%)	± 20.8
Number of ^{150}Nd nuclei (0.5%)	± 1.0

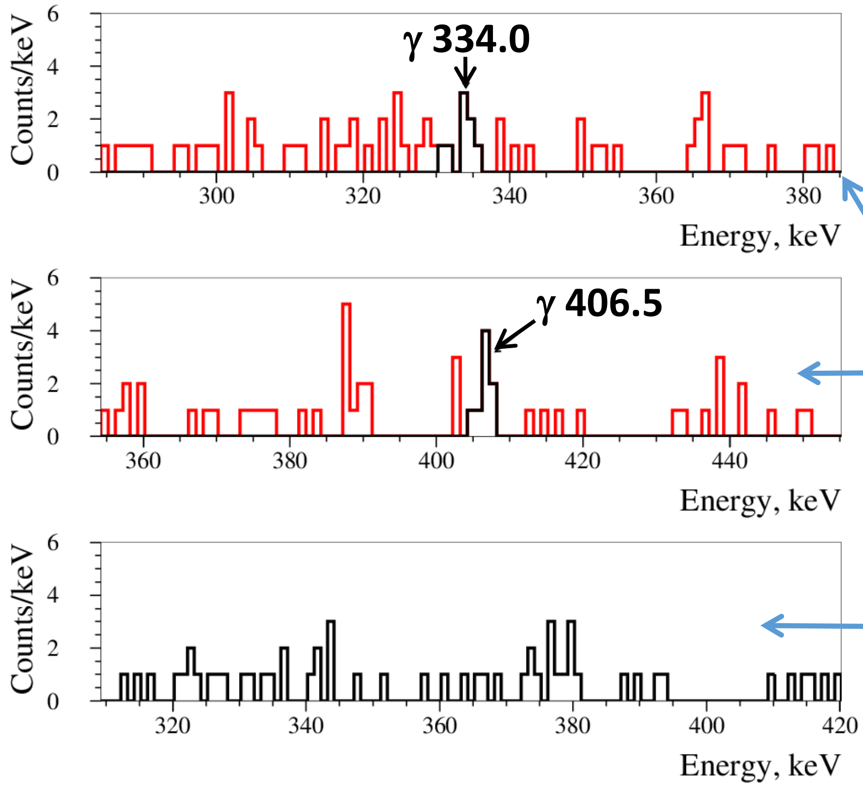
$$T_{1/2}^{406} = [17^{+17}_{-6} \pm 2\ (\mathrm{syst})] \cdot 10^{19}\mathrm{y}$$

Coincidence spectrum



- The two-dimensional energy spectrum of coincidences allows us to observe γ quanta emitted in the cascade (*left diagram*);
- The spectrum when the energy in one detector is fixed as (609 ± 5) keV (^{214}Bi, *top right*).
- The energy of one detector is fixed as (2615 ± 5) keV (^{208}Tl, *bottom right*).

Analysis of coincidences



The energy in one detector is fixed to the energy interval where γ quanta from the $^{150}Nd \rightarrow {}^{150}Sm$ (0^+, 740.5 keV) decay are expected:

406.5 keV ± 2.5×SD

334.0 keV ± 2.5×SD

A random coincidence background when energy of events in one of the detectors was taken as

375 keV ± 2.5×SD

$$S_{CC} = \mathbf{6.0}^{+3.3}_{-2.7}(\text{stat}) \pm \mathbf{0.9}(\text{syst}) \text{ counts}$$

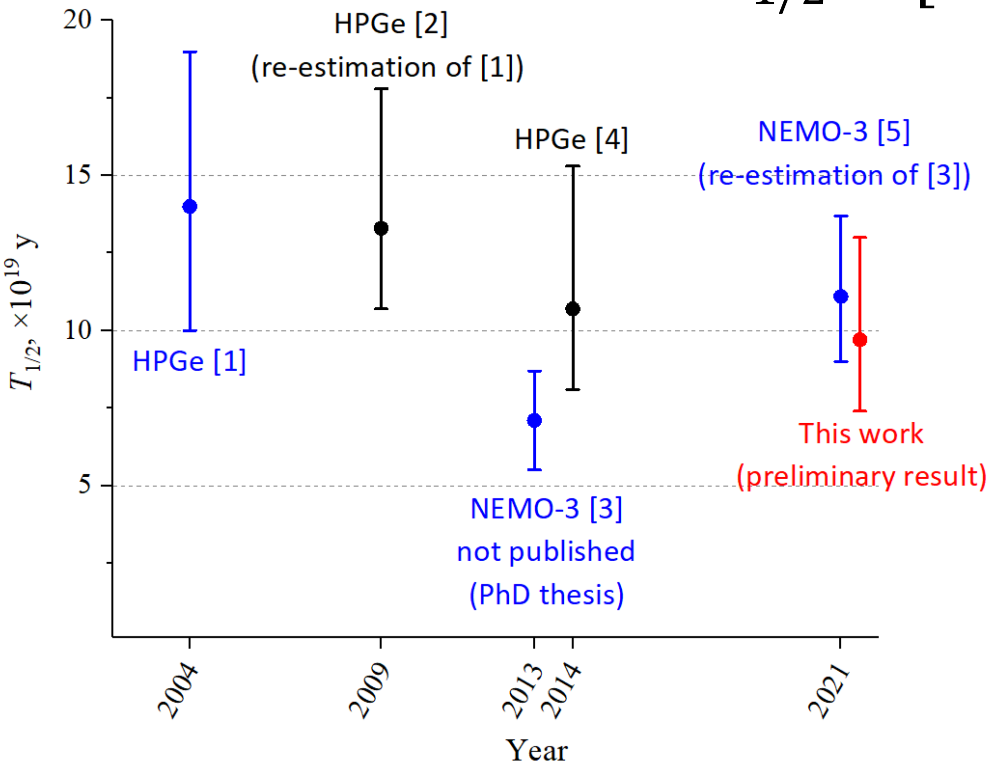
$$T^{CC}_{1/2} = \left[10^{+10}_{-5}(\text{stat}) \pm 2(\text{syst})\right] \cdot 10^{19}\text{y}$$

Combined value (334 keV and 406.5 keV)

$$T_{1/2}^{comb} = \frac{\ln 2 \cdot N \cdot t \cdot \sum \varepsilon_i}{\sum S_i\,(1+\alpha_i)}$$

$$\mathbf{T_{1/2}^{comb} = \left[9.7^{+2.9}_{-1.9} \pm 1.5\ (syst)\right] \cdot 10^{19} y}$$

$$T_{1/2}^{CC} = \left[10^{+10}_{-5}(\mathrm{stat}) \pm 2(\mathrm{syst})\right] \cdot 10^{19}\mathrm{y}$$



The experiment is in progress to improve the statistics

[1] A.S. Barabash et al., *Phys. Atom. Nucl.* **67** (2004) 1216.

[2] A.S. Barabash et al., *Phys. Rev. C* **79** (2009) 045501.

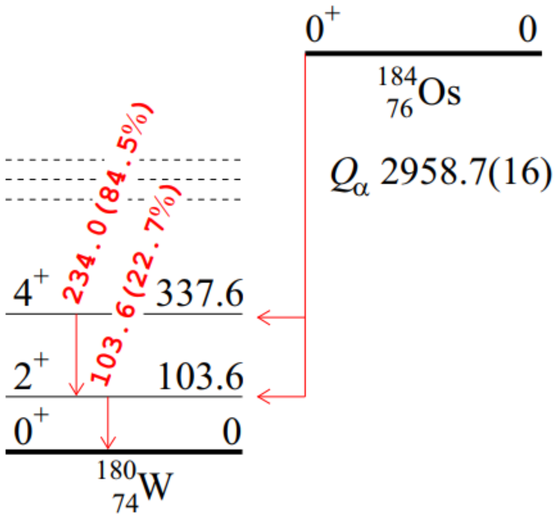
[3] S. Blondel, PhD thesis, LAL, Orsay, France (2013).

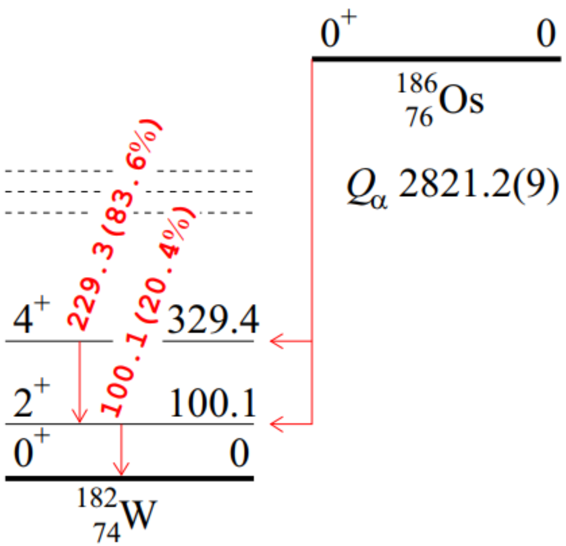
[4] M.F. Kidd et al., *Phys. Rev. C* **90** (2014) 055501.

[5] V. Tretyak, LXXI Int. Conf. “NUCLEUS-2021”, 20-25 Sep 2021, Book of Abstracts, Saint Petersburg (2021), p. 257

2. Search for α decay and 2β decay of naturally occurring osmium nuclides accompanied by γ quanta

α decay in naturally occurring Os isotopes

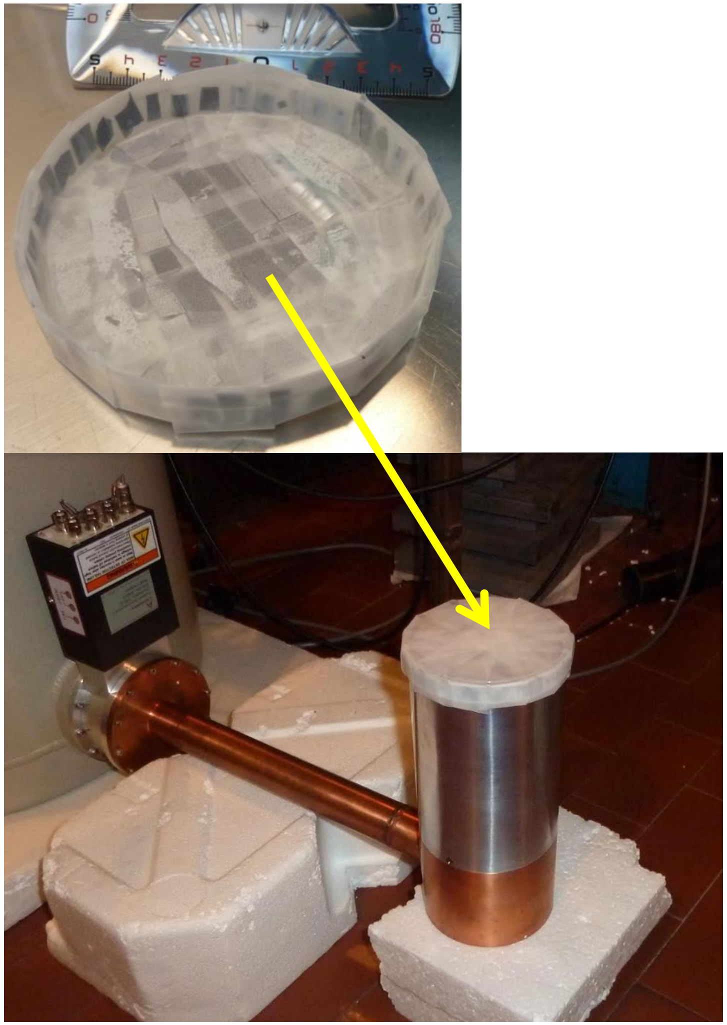




- All the 7 isotopes of natural Os are potentially unstable relative to α decay (A = 184, 186, 187, 188, 189, 190, 192)
- ^{184}Os ([1], geochemical in meteorites) and ^{186}Os ([2], direct observation) g.s.-g.s. transitions were observed
- ^{184}Os and ^{186}Os are prospective to search for α decay to the 1st excited states of daughters (experimental sensitivity is on the level of predictions).

[1] S. T. M. Peters *et al.*, Earth Planet. Sci. Lett. **391**, 69 (2014).
[2] V. E. Viola *et al.*, J. Inorg. Nucl. Chem. **37**, 11 (1975).

Experiment description



STELLA facility, LNGS (Italy) [1]

Os sample:

- 99.999% purity grade
- ingots obtained from osmium powder and used in the previous experiment [1] were cut into (0.8-1.3)-mm slices for this measurement
- mass of 117.96(2) g
- placed directly on the cryostat endcap of the 112.5-cm^3 BEGe detector (dead layer of 0.4 μm)

Passive shield made of radiopure copper (4-5 cm) and lead (20 cm)

Measurement time 15851 h (1.8 y)

[1] M. Laubenstein, Int. J. Mod. Phys. A **32** (2017) 1743002

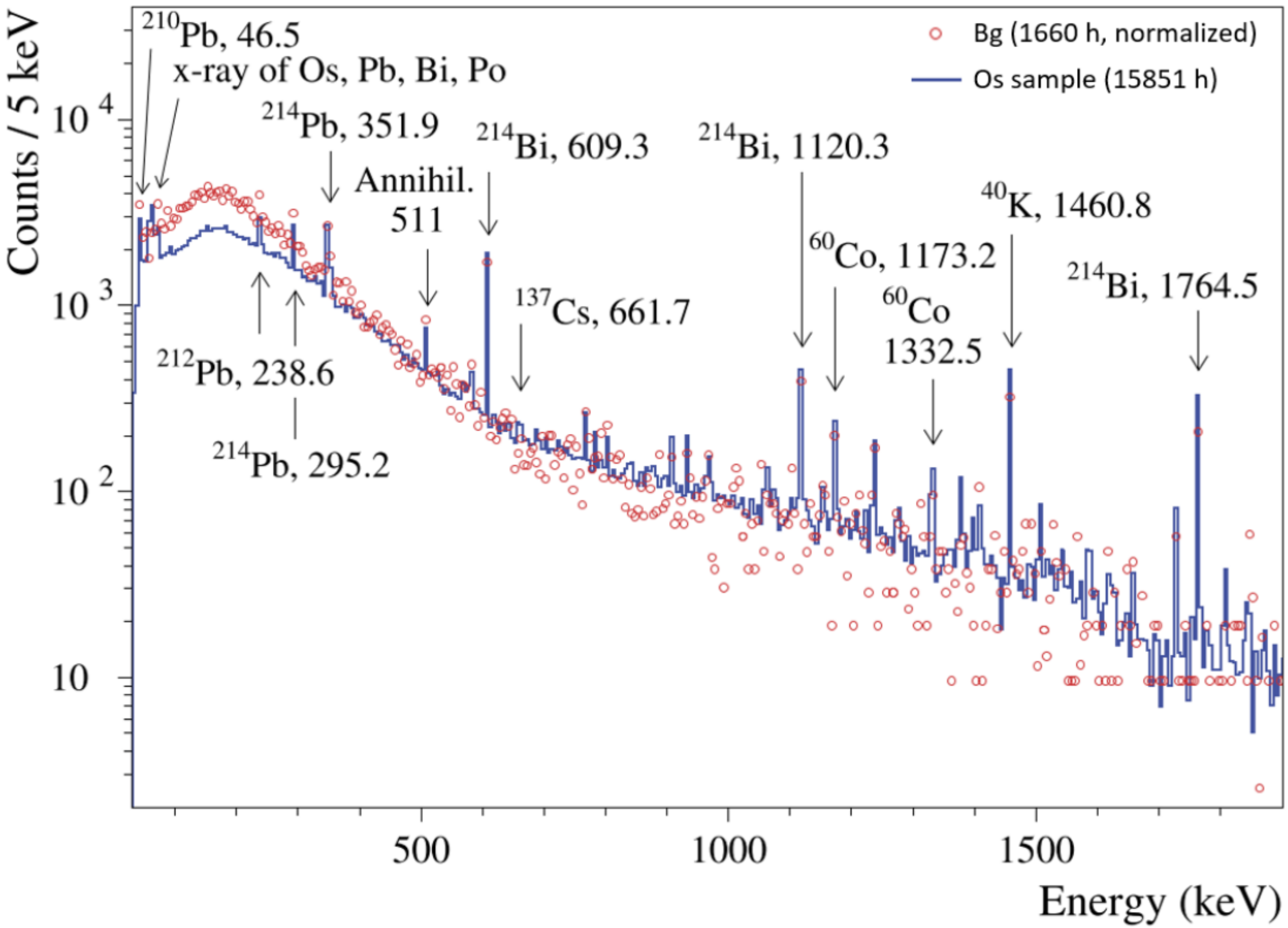
Isotopic composition of osmium

- John de Laeter Centre at Curtin University (Perth, Western Australia)
- Negative thermal ionization mass spectrometry (N-TIMS)
- Relative uncertainties for all the isotopes have been improved by 1-3 orders of magnitude (^{184}Os: 100% → 4.1%, ^{186}Os: 40.3% → 0.04%)

Isotope	δ (%) IUPAC [1]	δ (%) This work [2]	Number of nuclei in the sample
^{184}Os	0.02(2)	0.0170(7)	$6.35(26) \times 10^{19}$
^{186}Os	1.59(64)	1.5908(6)	$5.9405(25) \times 10^{21}$
^{187}Os	1.96(17)	1.8794(6)	$7.0182(25) \times 10^{21}$
^{188}Os	13.24(27)	13.253(3)	$4.9490(14) \times 10^{22}$
^{189}Os	16.15(23)	16.152(4)	$6.0316(18) \times 10^{22}$
^{190}Os	26.26(20)	26.250(8)	$9.8025(34) \times 10^{22}$
^{192}Os	40.78(32)	40.86(5)	$1.5258(19) \times 10^{23}$

[1] J. Meija et al., *Pure Appl. Chem.* **88** (2016) 293 [2] P. Belli et al., *Phys. Rev. C* **102** (2020) 102

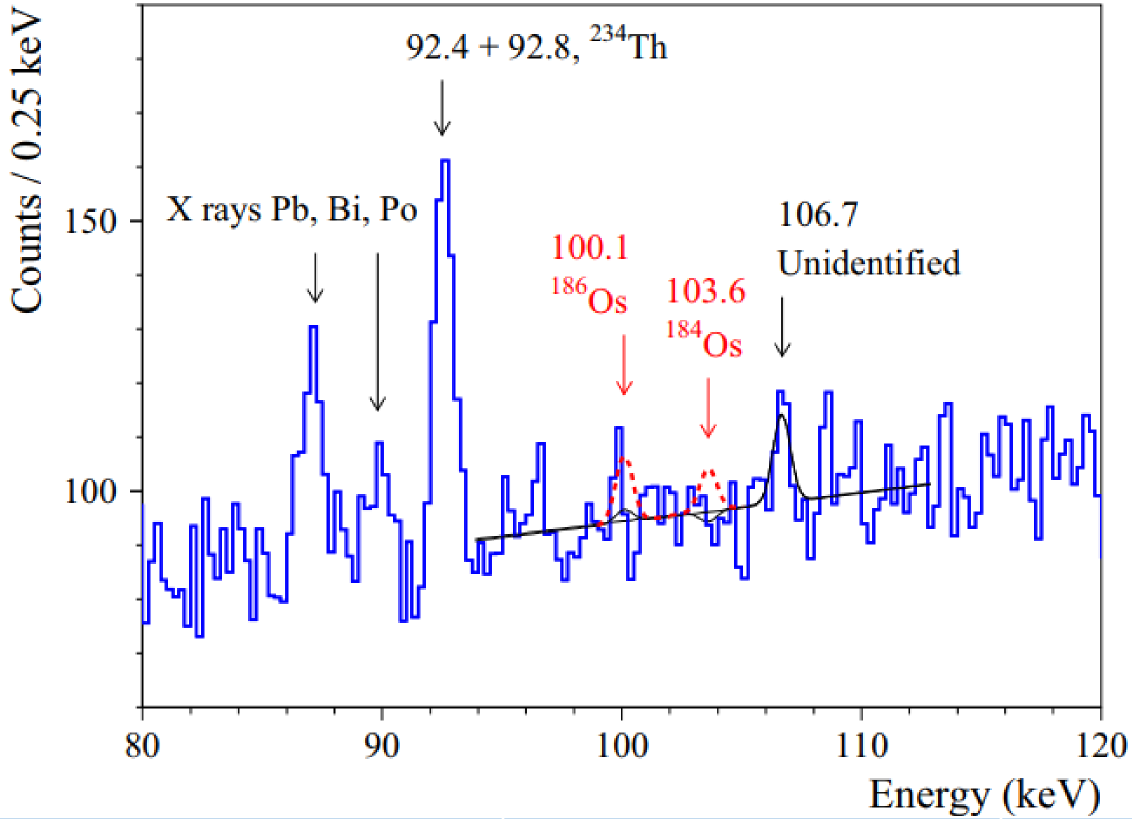
Os sample vs. background



Radioactive contamination of the Os sample

Decay chain	Radionuclide	Specific activity (mBq/kg)
	^{40}K	11 ± 4
	^{60}Co	$\leqslant 1.3$
	^{137}Cs	0.5 ± 0.1
^{232}Th	^{228}Ra	$\leqslant 6.6$
	^{228}Th	$\leqslant 16$
^{235}U	^{235}U	$\leqslant 8.0$
	^{231}Pa	$\leqslant 3.5$
	^{227}Ac	$\leqslant 1.1$
^{238}U	^{238}U	$\leqslant 35$
	^{226}Ra	$\leqslant 4.4$
	^{210}Pb	$\leqslant 180$

^{184,186}Os α decay to the 1st excited levels of ^{180,182}W



Transition	lim $T_{1/2}$, y (90% C.L.)	Predicted $T_{1/2}$, y
$^{184}Os \rightarrow {}^{180}W(2^+, 103.6\ keV)$	8.8×10^{15}	$(0.6 - 2.9) \times 10^{15}$
$^{186}Os \rightarrow {}^{182}W(2^+, 100.1\ keV)$	4.4×10^{17}	$(0.3 - 2.2) \times 10^{17}$

The limits substantially exceed the theoretical predictions!

Application of systematics [1]

Source	relative systematic uncertainties	
	^{184}Os	^{186}Os
Detection efficiency	0.098	0.118
Interval of fit	0.076	0.065
Isotopic abundance	0.041	0.0004
Total relative systematic error (σ_r)	0.131	0.135

$$\lim S \to \lim S' = \lim S \times a$$

$$a = [1 + (\lim S - S) \times \sigma_r^2/2]$$

where σ_r is a relative systematic uncertainty of the peak area *S*.

Corrected $T_{1/2}$ limits:

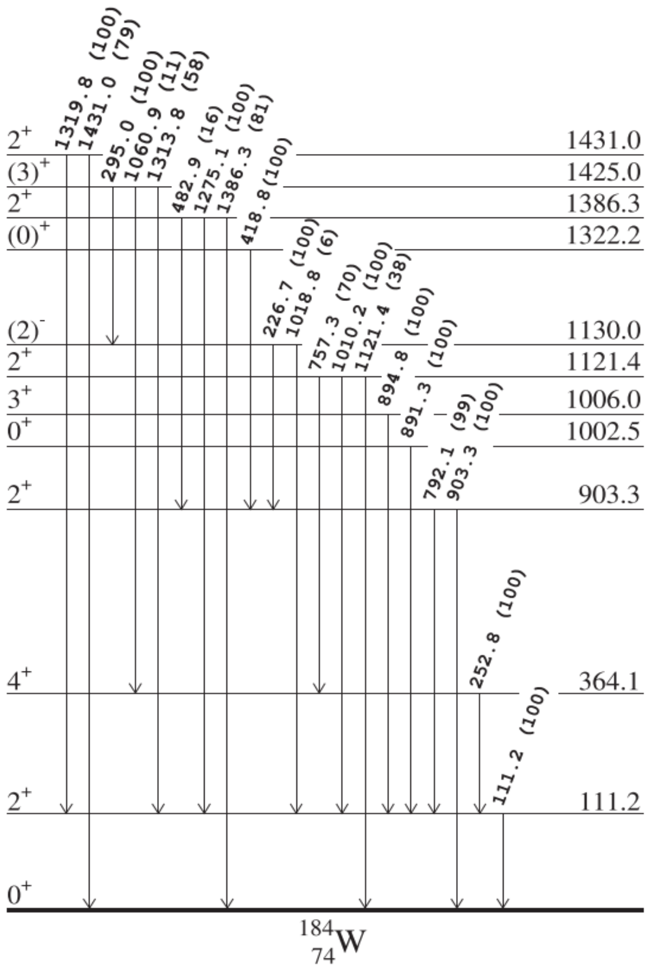
$$\lim T_{1/2}(^{184}\mathrm{Os}) = 6.8 \times 10^{15}\ \mathrm{y},\ \lim T_{1/2}(^{186}\mathrm{Os}) = 3.3 \times 10^{17}\ \mathrm{y}$$

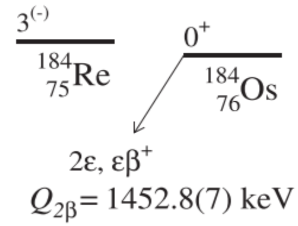
[1] R.D. Cousins and V.L. Highland, *Nucl. Instrum. Meth. A* **320** (1992) 331

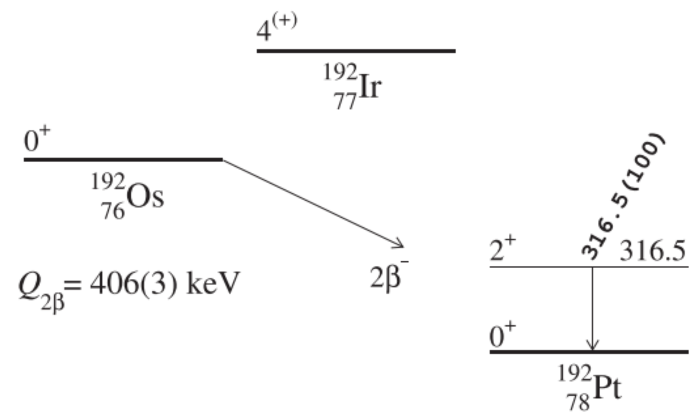
Summary

Nuclide	δ, %	Daughter level, energy in keV	Q_α, keV (g.s.-g.s.)	$T_{1/2}$, y	
				predictions	This work
^{184}Os, 0^+	0.0158(11)	^{180}W, 2^+, 103.6	2958.7(16)	$(1.3 – 2.9)\times10^{15}$	**$\geq 6.8 \times 10^{15}$**
		^{180}W, 4^+, 337.6		$(0.09 – 2.5)\times10^{19}$	$\geq 4.6 \times 10^{16}$
^{186}Os, 0^+	1.5908(6)	^{182}W, 2^+, 100.1	2821.2(9)	$(0.3 – 2.2)\times10^{17}$	**$\geq 3.3 \times 10^{17}$**
		^{182}W, 4^+, 329.4		$(0.07 – 2.9)\times10^{21}$	$\geq 6.0 \times 10^{18}$
^{187}Os, $1/2^-$	1.8794(6)	^{183}W, $3/2^-$, 46.5	2721.7(9)	$1.6\times10^{17} – 4.4\times10^{20}$	$\geq 3.2 \times 10^{15}$
		^{183}W, $5/2^-$, 99.1		$9.1\times10^{17} – 2.8\times10^{21}$	$\geq 1.9 \times 10^{17}$
^{188}Os, 0^+	13.253(3)	^{184}W, 2^+, 111.2	2143.2(9)	$(0.1 – 2.9)\times10^{29}$	$\geq 3.3 \times 10^{18}$
		^{184}W, 4^+, 364.1		$8.9\times10^{33} – 1.9\times10^{36}$	$\geq 5.0 \times 10^{19}$
^{189}Os, $3/2^-$	16.152(4)	^{185}W, $3/2^-$, g.s.	1976.1(9)	$3.1\times10^{29} – 2.4\times10^{34}$	$\geq 3.5 \times 10^{15}$
		^{185}W, $1/2^-$, 23.5		$3.2\times10^{30} – 1.8\times10^{35}$	$\geq 3.5 \times 10^{15}$
		^{185}W, $5/2^-$, 65.9		$3.1\times10^{31} – 2.1\times10^{36}$	$\geq 7.6 \times 10^{17}$
^{190}Os, 0^+	26.250(8)	^{186}W, 2^+, 122.6	1375.8(12)	$1.6\times10^{51} – 1.1\times10^{54}$	$\geq 1.2 \times 10^{19}$
		^{186}W, 4^+, 396.5		$1.6\times10^{65} – 5.8\times10^{69}$	$\geq 8.6 \times 10^{19}$
^{192}Os, 0^+	40.78(32)	^{188}W, 0^+, g.s.	361(4)	$1.4\times10^{140} – 1.7\times10^{153}$	$\geq 5.8 \times 10^{18}$
		^{188}W, 2^+, 143.2		$9.9\times10^{190} – 1.6\times10^{215}$	$\geq 2.7 \times 10^{19}$

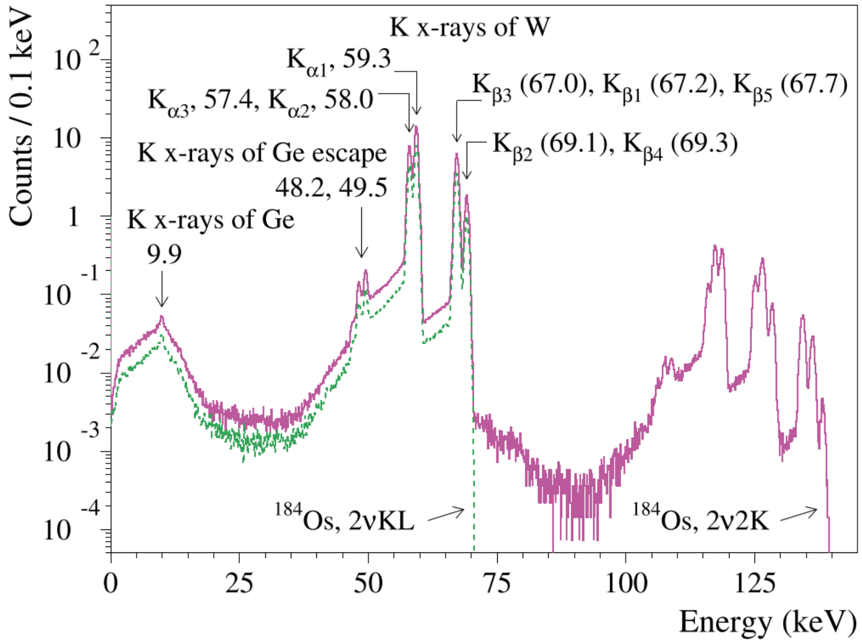
2β processes in Os nuclides



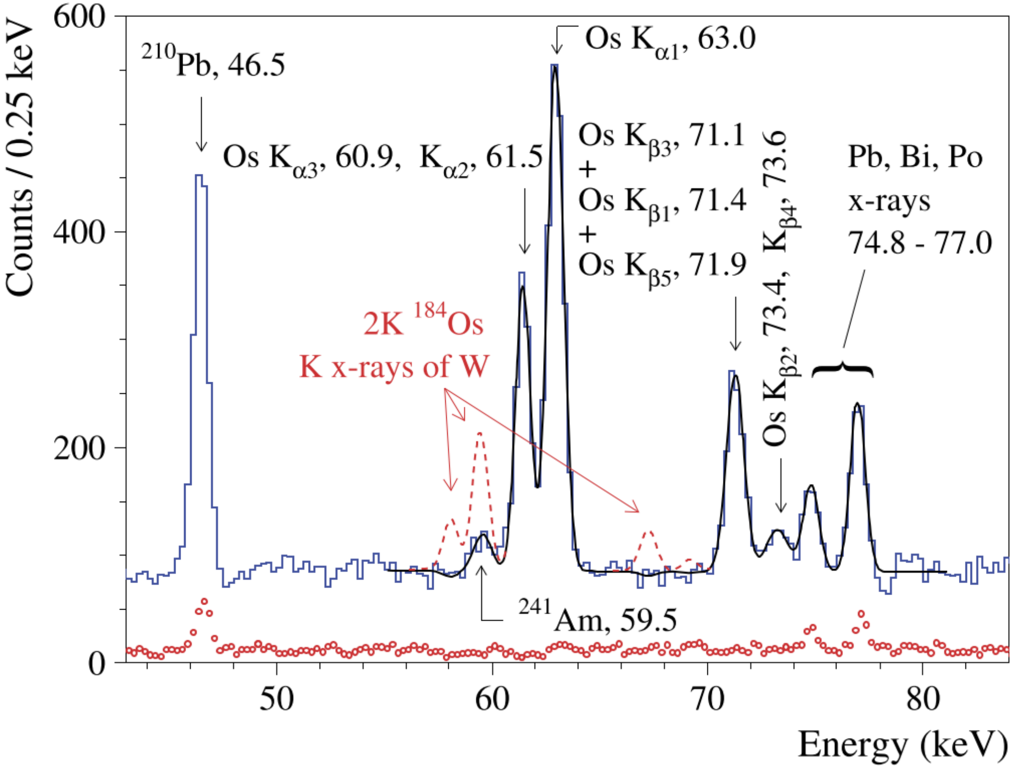




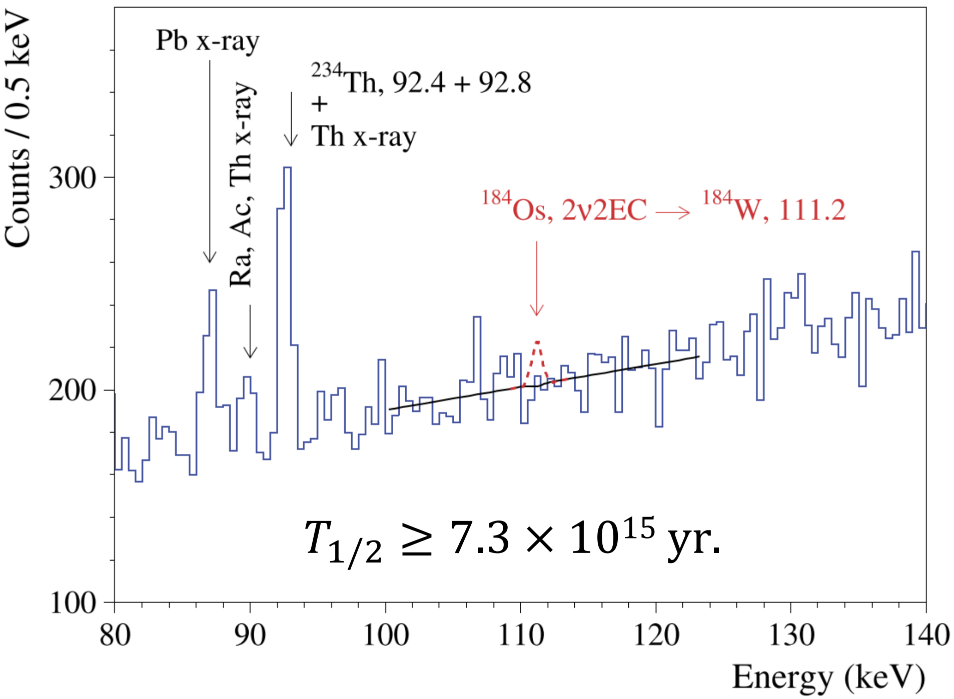
2ν2K and 2νKL decays of ^{184}Os

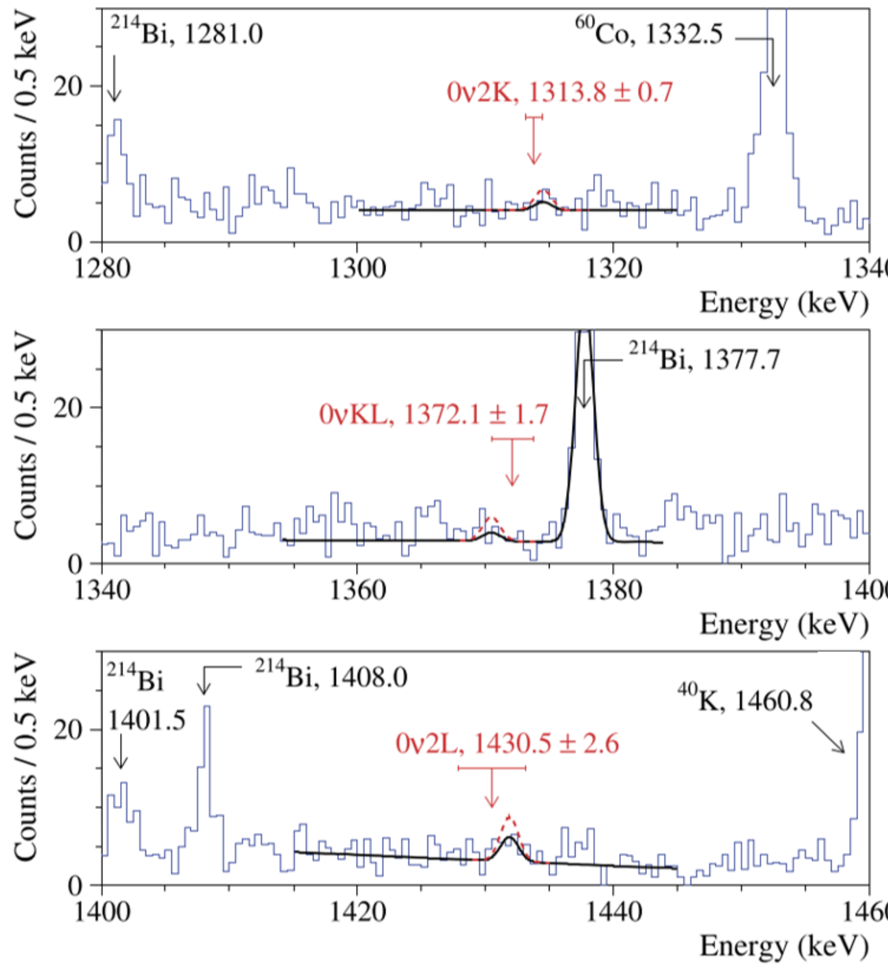


Transition	Final level of ^{184}W	Lim $T_{1/2}$, y
2ν2K	g.s.	3.0×10^{16}
	2^+, 111.2 keV	3.6×10^{16}
2νKL	g.s.	2.0×10^{16}
	2^+, 111.2 keV	2.4×10^{16}
0νKL	2^+, 111.2 keV	1.9×10^{16}



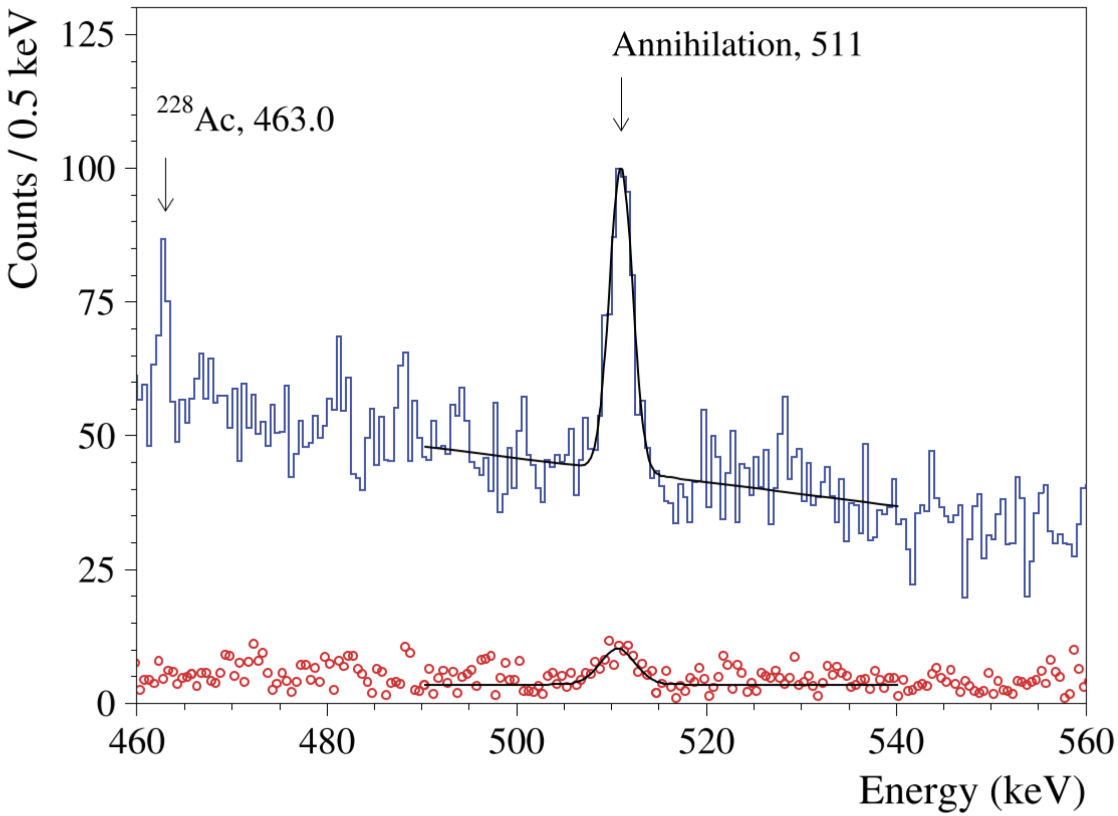
2EC decays of ^{184}Os to 111.2-keV daughter level





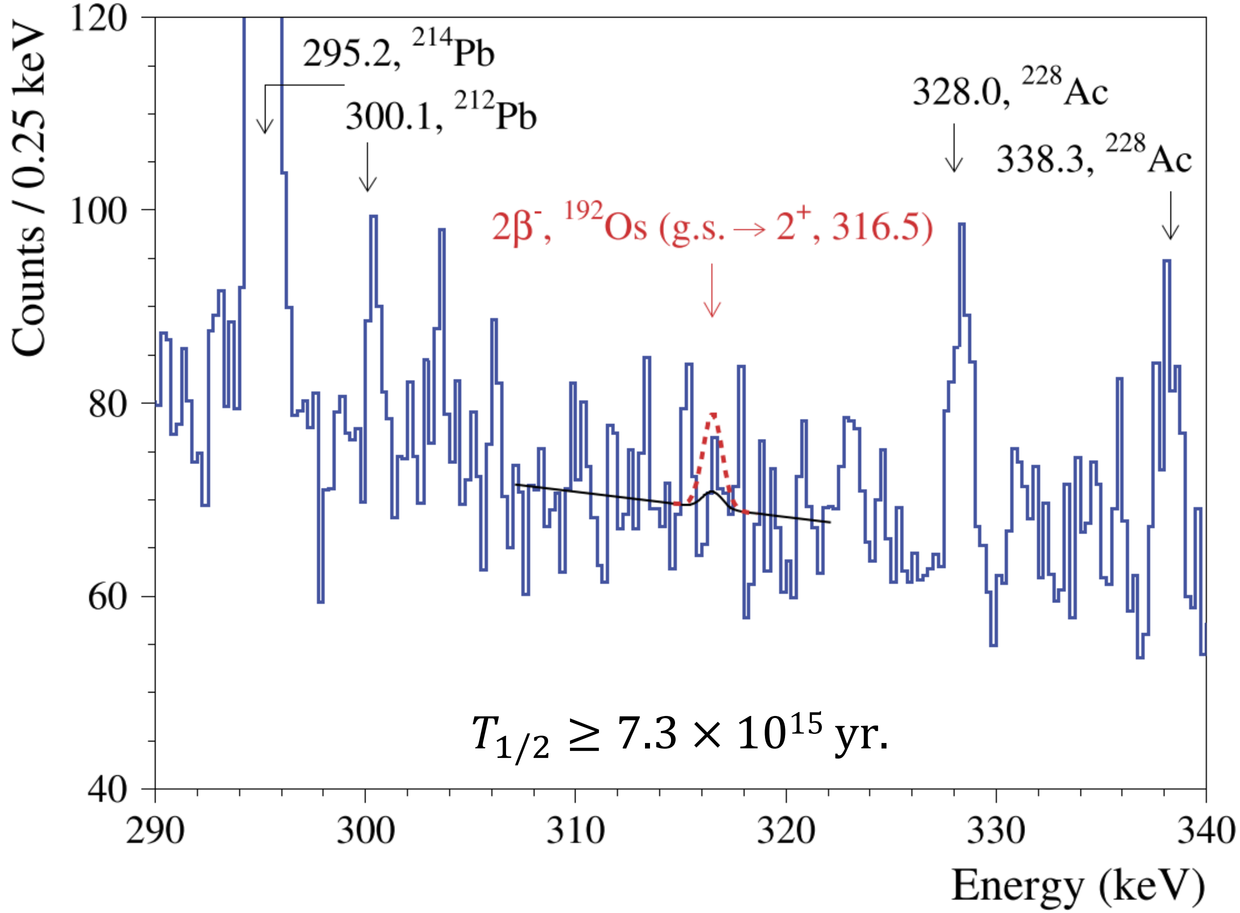
Transition	Final level of ^{184}W	Lim $T_{1/2}$, y
2ν2EC	111.2 keV	7.3×10^{15}
0ν2K	g.s.	1.6×10^{17}
0νKL	g.s.	1.3×10^{17}
0ν2L	g.s.	7.3×10^{16}

ECβ⁺ decay of ^{184}Os



Transition	Final level of ^{184}W	Lim $T_{1/2}$, y
2νECβ⁺	g.s.	1.0×10^{17}
	2⁺, 111.2 keV	1.0×10^{17}
0νECβ⁺	g.s.	1.0×10^{17}
	2⁺, 111.2 keV	9.9×10^{16}

2β⁻(2ν+0ν) decay of ^{192}Os (to 316.5-keV level)



Comparison with previous result (1)

Process, daughter level	E_γ, keV	$T_{1/2}$, yr	
		This work [1]	Previous [2]
$^{184}Os \rightarrow {}^{184}W$			
2ν2K – g.s.	57–69	**≥ 3.0 × 10^{16}**	≥ 1.9 × 10^{14}
2νKL – g.s.	57–69	**≥ 2.0 × 10^{16}**	–
2ν2K – 2^+, 111.2	57–69	**≥ 3.6 × 10^{16}**	≥ 3.1 × 10^{15}
2νKL – 2^+, 111.2	57–69	**≥ 2.4 × 10^{16}**	≥ 3.1 × 10^{15}
2ν2EC – 2^+, 111.2	111.2	**≥ 7.3 × 10^{15}**	≥ 3.1 × 10^{15}
2ν2EC – 2^+, 903.3	903.3	**≥ 2.0 × 10^{17}**	≥ 3.2 × 10^{16}
2ν2EC – 0^+, 1002.5	891.3	≥ 2.8 × 10^{17}	≥ 3.8 × 10^{17}
2ν2EC – 2^+, 1121.4	757.3	**≥ 1.0 × 10^{17}**	≥ 6.9 × 10^{16}
2νKL – (0^+), 1322.2	903.3	**≥ 1.7 × 10^{17}**	–
2ν2L – 2^+, 1386.3	1275.1	**≥ 3.0 × 10^{16}**	–
2ν2L – $(3)^+$, 1425.0	903.3	**≥ 8.4 × 10^{16}**	–
2ν2L – 2^+, 1431.0	1319.8	**≥ 4.4 × 10^{16}**	–
0ν2K – g.s.	1313.8(7)	≥ 1.6 × 10^{17}	≥ 2.0 × 10^{17}
0νKL – g.s.	1372.1(17)	≥ 1.3 × 10^{17}	≥ 1.3 × 10^{17}
0ν2L – g.s.	1430.5(26)	≥ 7.3 × 10^{16}	≥ 1.4 × 10^{17}

Process, daughter level	E_γ, keV	$T_{1/2}$, y	
		This work [1]	Previous [2]
$^{184}Os \rightarrow {}^{184}W$			
0ν2K – 2^+, 111.2	1202.6(7)	≥ 7.6 × 10^{16}	≥ 3.3 × 10^{17}
0νKL – 2^+, 111.2	57–69	**≥ 1.9 × 10^{16}**	–
0ν2EC – 2^+, 903.3	903.3	**≥ 1.7 × 10^{17}**	≥ 2.8 × 10^{16}
0ν2EC – 0^+, 1002.5	310.6–312.0	≥ 2.1 × 10^{17}	≥ 3.5 × 10^{17}
0ν2EC – 2^+, 1121.4	757.3	**≥ 9.4 × 10^{16}**	≥ 6.4 × 10^{16}
0νKL – (0^+), 1322.2	903.3	**≥ 1.7 × 10^{17}**	≥ 2.8 × 10^{16}
0ν2L – 2^+, 1386.3	1275.1	≥ 3.0 × 10^{16}	≥ 6.7 × 10^{16}
0ν2L – $(3)^+$, 1425.0	903.3	**≥ 8.4 × 10^{16}**	–
0ν2L – 2^+, 1431.0 resonant	1319.8	≥ 4.4 × 10^{16}	≥ 8.2 × 10^{16}
2νECβ^+ – g.s.	511	**≥ 1.0 × 10^{17}**	≥ 2.5 × 10^{16}
2νECβ^+– 2^+, 111.2	511	**≥ 1.0 × 10^{17}**	≥ 2.5 × 10^{16}
0νECβ^+ – g.s.	511	**≥ 1.0 × 10^{17}**	≥ 2.5 × 10^{16}
0νECβ^+ – 2^+, 111.2	511	**≥ 9.9 × 10^{16}**	≥ 2.4 × 10^{16}
$^{192}Os \rightarrow {}^{192}Pt$			
(2ν+0ν)2β^- – 2^+ 316.5	316.5	**≥ 2.0 × 10^{20}**	≥ 5.3 × 10^{19}

[1] Belli et al., *J. Phys. G* **48** (2021) 085104
[2] Belli et al. *Eur. Phys. J. A* **49** (2013) 24

Conclusions

- 2β decay of ^{150}Nd to the first 0^+ excited state of ^{150}Sm has been investigated with ~2.4-kg Nd_2O_3 sample by using low-background 4-crystal HPGe γ-spectrometer. The half-life value has been obtained after 4.5 yr. of data taking to be

$$T_{1/2} = [9.7^{+2.9}_{-1.9}(\text{stat}) \pm 1.5\ (\text{syst})] \cdot 10^{19}\text{y (preliminary)}.$$

  The measurement is in progress to increase the statistics.

- α and double-β processes in Os naturally occurring isotopes were searched for over 1.8 yr. using low-background 112-cm^3 BEGe detector and 118-g sample of osmium.
- The half-life limits for $^{184,186}Os$ relative to α decay to the 1st excited states of daughters are measured to be substantially higher than theoretical predictions for these transitions.
- New or improved half-life limits on most of the 2β decay channels of ^{184}Os have been set at the level of $10^{16} - 10^{17}$ y at 90% C.L. The half-life limit on $2\beta^-$ decay of ^{192}Os to the first excited level of ^{192}Pt has been 4 times increased compared to the previous result.
- The next stage of the experiment is in progress with a sample placed directly on the Ge crystal inside the cryostat to improve detection efficiency.