Search for long-lived superheavy ekatungsten (Sg – Seaborgium) with radiopure enriched ¹¹⁶CdWO₄ crystal scintillators

<u>O.G. Polischuk¹</u>, P. Belli^{2,3}, R. Bernabei^{2,3}, F. Cappella^{4,5}, V. Caracciolo⁶, R. Cerulli^{2,3}, F.A. Danevich¹, A. Incicchitti^{4,5}, D.V. Kasperovych¹, V.V. Kobychev¹, D.V. Poda^{1,7}, V.I. Tretyak¹

- ¹ Institute for Nuclear Research, Kyiv, Ukraine
- ² Dipartimento di Fisica, Universita di Roma "Tor Vergata", Rome, Italy
- ³ INFN sezione di Roma "Tor Vergata", Rome, Italy
- ⁴ INFN, sezione di Roma, Rome, Italy
- ⁵ Dipartimento di Fisica, Universita di Roma "La Sapienza", Rome, Italy
- ⁶ INFN, Laboratori Nazionali del Gran Sasso, Assergi (AQ), Italy
- ⁷ Centre de Sciences Nucleaires et de Sciences de la Matiere, Orsay, France

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Island of stability



It is expected that only the edge of the 'island of stability' has been reached to date in laboratory conditions, long-lived SHEs were probably produced in explosive stellar events by a sequence of rapid neutron captures (r-processes, but the question is – if in sufficient amount?) and β decays.

In 2006 scientists from JINR(Russia) led by Oganessian announced first element with a long-lived number 114 which means the experimental confirmation of the existence of the "island of stability".

- [1] Nature 231(1971)103
- [2] Phys. At. Nucl. 72(2009)1026
- [3] Eur. Phys. J. A 48(2012)122
- [4] Astron. Lett. 39(2013)150
- [5] Oganessian et al. Phys. Rev. C. 70(2004)064609

Superheavy elements (SHE) in nature

Z≥ 104 and A≥ 250 [1];
formed in fusion reaction [2] or r-processes;
T_{1/2} – from μs to hours (with number of neutrons near the magic number 184 are expected to be longer);

SHE with Z= 102 – 107 lie in the interval 7.8 – 10.6 MeV [3];

- strong increase in the stability of nuclei near the magic numbers Z = 114 and N = 184, which could ⁸⁰ lead to the existence of stability islands (due to the increased stability provided by shell effects). ⁶⁰

Could decay through:

- emission of β^- particles or by EC (or β^+ decay); 20
- α decays followed by spontaneous fission [4];
- cluster decay [5]

[1] Nucleonics 15 (1957) 122;

- [2] Rev. Mod. Phys. 72 (2000) 733; J. Phys. G 34 (2007) R165
- [3] At. Data Nucl. Data Tables 98 (2012) 1096
- [4] Annu. Rev. Nucl. Part. Sci. 63 (2013) 383
- [5] Prog. Part. Nucl. Phys. 58 (2007) 292



The possibility of the existence of superheavy nuclei is of significant importance for the understand ing of the properties of nuclear matter. 3

Studying of long-lived isotopes of a SHE in natural Au by ISPMS

The closest chemical homologue for Au among SHE is Rg (Roentgenium) with Z = 111 (if exist in nature!).

The accurate mass measurements (using a high resolution inductively coupled plasma- sector field mass spectrometer (ICP-SFMS)) for masses 254 and 259–269 was performed.

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Mass	Fig.	No. of			Mass of
no.	no.	events	$P_{\rm acc.}$	$M_{ m c.m.}^{ m exp.a}$	Rg isotope ^b
261	2(a)	6	$8 imes 10^{-7}$		
261	2(b)	22(18)	$3 \times 10^{-6 \mathrm{c}}$	261.134^{d}	261.154
265	3(a)	4	2×10^{-6}		
265	3(b)	10	3×10^{-9}	265.154	265.151

Evidence for the existence of isotopes with masses that fit the predictions for the masses of 261 Rg and 265 Rg was obtained. The predicted g.s. half-lives of these Rg isotopes are of the order of 1 µs. This suggests that the observed events are due to long-lived Rg isomers.

Search for SHE in nature with accelerator mass spectrometry

Problems of the artificial creation in laboratory :

- + production of isotopes of nuclei near the stability island
- + AMS does not have to deal with molecular background, unlike other mass spectrometry methods such as ICPMS
- the extremely small cross sections;
- the lack of stable target-projectile combinations neutron-rich enough to reach the theoretical island of stability;
- half-lives of SHEs could be so short that their abundance in the samples has dropped below detection limits in the time since their synthesis until today;
- should be synthesized in sufficient amounts in the rapid neutron capture process (then could be still present in nature)
- discovery without confirmation: different approach couldn't reach results of each other (for example, SHE with Z = 122 and A = 292 in a sample of natural thorium by using inductively coupled plasma mass spectrometry (ICPMS) could not be confirmed by using accelerator mass spectrometry (AMS) and is thus doubtful);
- ✓ It is favorable to use a sample material consisting of several possibly chemically homologous elements for a wide-spread search for SHEs in nature.

Phys. Rev. C 85 (2012) 024315 + Phys. Rev. C 83 (2011) 015801 + Phys. Rev. C 83 (2011) 065806 Int. J. Mod. Phys. E 19(2010) 131

Search for SHE in Pt with accelerator mass spectrometry



A total of 14 different masses in the range 292 < A < 310 were scanned with AMS.

The use of raw platinum allowed to scan for several SHEs in one sample material, (it contains different possible chemical homologues to SHEs: **Ru, Rh, Pd, Os, Ir, Pt**).

Ash	Sample	Ref. isotope	$m_{\rm ad}(\%)$	$A_{\rm ad}$ (%)	930	S_{ul} (%)	$S_{\rm SHE}(\%)$	7 (nA)	Time (h)	$\epsilon~(\%)$	Rate (Hz)	AHE ev
292	Os	182Os ⁸⁺	41.37	41.0	11+	17.1	17.8	11.6	25.6	18.0	31	.0
292	Ou.	142Os74	41.37	41.0	10+	14.4	19.1	11.1	10.6	26.4	0.01	0
292	raw Pt	188 Pt ==	2.35	0.70	10+	14.7	19.1	4.9	2.3	20.0	9	0
293	raw Pt	105pt7+	2.35	0.70	11+	14.7	18.4	8.7	3.4	31.5	2	0
294	raw Pt	198 Pt 24	2.35	0.70	11+	14.7	18,5	2.8	9.7	31.5	17	0
295	nw Pt	186Pt2+	2.35	0.70	10+	14.7	18.9	6.8	7.1	31.0	6	0
297	raw Pt	132 pts+	2.35	0.70	11+	16.7	17.7	5.0	5.0	12.4	6	0
298	PbF ₂	366Pb7+	52.59	52.4	11+	18.3	16.2	13.1	5.2	19.6	11	0
299	raw Pt	100Pt++	2.35	0.70	12+	16.7	16.0	2.6	3.3	12.2	81	0
300	raw Pt	tao blav	2.35	0.70	12+	16.7	16.1	5.7	2.2	16.4	7	0
301	raw Pt	100 Pt 0+	2.35	0.70	12+	16.7	16.1	3.0	1.8	12.5	1	0
302	raw Pt	(mpt++	2.35	0.70	13+	16.7	14.0	6.3	2.5	20.5	50	0
304	raw Pt	196 P18+	2.35	0.70	12+	16.7	16.1	2.0	4.2	12.5	2	0
306	new Pt	198 P1 ⁸⁺	2.35	0.70	12+	16.7	17.6	2.8	3.6	12.8	8	0
308	raw Pt	tesbfa+	2.35	0.70	12+	16.7	16.1	1.1	1.7	12.9	136	0
310	raw Pt	198Pt2+	2.35	0.70	11+	14.7	18.0	4.9	62	29.1	33	0

²⁹⁸114 (by Oganessian) was conducted in its possible chem. homologue lead
There were no SHE events recorded for any of the mass settings → upper limits on their abundances in the sample materials are of the order 10⁻¹⁴ – 10⁻¹⁶

Ratio	Sample	Upper limit $\left[\frac{\text{atoms}}{\text{atoms}}\right]$	Upper limit $\left[\frac{g}{g}\right]$
²⁹² Hs/Os	Os	2.0×10^{-15}	3.0×10^{-15}
²⁹² X/raw Pt	raw Pt	9.4×10^{-16}	4.8×10^{-15}
²⁹³ Mt/Ir	raw Pt	3.6×10^{-14}	5.4×10^{-14}
²⁹³ X/raw Pt	raw Pt	2.4×10^{-16}	1.2×10^{-15}
²⁹⁴ Ds/Pt	raw Pt	2.7×10^{-15}	4.0×10^{-15}
²⁹⁴ X/raw Pt	raw Pt	2.6×10^{-16}	1.3×10^{-15}
²⁹⁵ Rg/Au	raw Pt	4.1×10^{-14}	6.1×10^{-14}
²⁹⁵ X/raw Pt	raw Pt	1.5×10^{-16}	7.3×10^{-16}
²⁹⁷ X/raw Pt	raw Pt	9.9×10^{-16}	4.9×10^{-15}
²⁹⁸ Uuq/Pb	PbF ₂	1.8×10^{-14}	2.6×10^{-14}
²⁹⁹ X/raw Pt	raw Pt	3.2×10^{-15}	1.6×10^{-14}
³⁰⁰ X/raw Pt	raw Pt	1.6×10^{-15}	8.2×10^{-15}
³⁰¹ X/raw Pt	raw Pt	4.9×10^{-15}	2.5×10^{-14}
³⁰² X/raw Pt	raw Pt	1.2×10^{-15}	6.1×10^{-15}
³⁰⁴ X/raw Pt	raw Pt	3.2×10^{-15}	1.6×10^{-14}
³⁰⁶ X/raw Pt	raw Pt	2.4×10^{-15}	1.2×10^{-14}
308 X/raw Pt	raw Pt	1.4×10^{-14}	7.2×10^{-14}
³¹⁰ X/raw Pt	raw Pt	2.6×10^{-16}	1.4×10^{-15}

Search for SHE in Galactic Cosmic Rays

6000 nuclei with Z>55 in galactic cosmic rays has been obtained in the **OLIMPIYA** project. **3** SHE with 105 < Z < 130 have been detected.

Their detection in nature could confirms theoretical predictions and justifies efforts for their synthesis under terrestrial conditions



The abundance of elements obtained in studies with aerostats, satellites, and meteorites

OLIMPIYA project



Search for the abundance of nuclei (with Z > 86) in cosmic rays is the extremely low flux of these nuclei (1-2 nuclei/m²/yr) with solid state track detectors, where particles are detected in terms of radiation damage induced by heavy and SHE from CR in olivine crystals from meteorites

Eagle Station meteorite sample used in the OLIMPIYA project



Distribution of the number of the detected and identified superheavy nuclei

Potentiality of scintillators to search for SHE

Searching for high energy α 's from the decay of natural SHE (or its daughters), embedded in a detector:

- 1) $ZnWO_4$ for superheavy eka-W (Z = 106)
- 2) $CdWO_4$ for superheavy eka-W (Z = 106)
- 3) $BiGe_3O_{12}$ (BGO) scintillators or scintillating bolometers for superheavy eka-Bi (Z = 115)
- 4) $PbWO_4$ for superheavy eka-Pb
- 5) NaI(Tl) for eka-Tl

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6) .....
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+ good PSD allows so select alpha events with high energy (> 8 MeV)

+ possibility to reject Bi-Po events, pile-ups...

 – can't distinguish a specific isotope (e.g. eka-W or eka-Bi) if alternative explanations (of events with shape of scintillation signal typical for α 's and high energy) will be absent, this would be an indication on presence of SHE

Decay of superheavy Sg (Z = 106)

Chemical properties of Sg are similar to those of W [1-3] and one could expect that long-lived Sg follows W in the processes of chemical separation and growth of the crystals, and could be present at some amount in the detectors.

- 1) Sg could decay through
- β^- channel (\Rightarrow Z = 107)
- $\operatorname{EC}/\beta + (\rightarrow Z = 105)$
- α decay (\rightarrow Z = 104) with "low-energy" α 's (4 6 MeV $\cong \alpha$'s from U/Th chains)

2) The created nucleus (or one of its daughters) decays with emission of high energy α particle (Q > 8 MeV)

3) If these α 's live long enough (seconds or larger), they could to be registered outside by data acquisition system. The theoretical predictions [4–6] also confirm this assumption.

[1] Nature 388 (1997) 55
 [2] J. Chem. Phys. 138 (2013) 174301
 [3] Science 345 (2014) 1491
 [4] At. Data Nucl. Data Tables 94 (2008) 781
 [5] Phys. Rev. C 81 (2010) 034613
 [6] Phys. Rev. C 86 (2012) 014322

SHE with ZnWO₄

Laboratori Nazionali del Gran Sasso (INFN, Italy)



 $ZnWO_4$ (699 g) was produced in the Institute for Scintillation Materials (ISMA, Kharkiv, Ukraine) from crystal ingots grown in platinum crucibles by the Czochralski method.

It was investigated in low background measurements during 2130 h at LNGS

We looked for high energy α particles (Q > 8 MeV)

Pulse-shape discrimination

The optimal filter method proposed by E. Gatti and F. De Martini [1] was applied



Shape indicator (SI):

$$SI = \frac{\sum f(t_k) \times P(t_k)}{\sum f(t_k)}$$

 $f(t_k)$ is a digital amplitude of a signal at the time channel t_k ; $P(t_k)$ is a weight function

$$P(t_k) = \frac{\left|f_{\alpha}(t) - f_{\gamma}(t)\right|}{f_{\alpha}(t) + f_{\gamma}(t)}$$

 $f_a(t)$ and $f_{\gamma}(t)$ are digital amplitudes of reference α and γ/β signals, respectively

[1] Proceedings of the Conference on Nuclear Electronics Vol. II (International Atomic Energy Agency, Vienna, 1962), p. 265.

Energy spectra of events selected by PSD



The energy distributions of the β particles (γ quanta) and α particles selected by applying the PSD. In the inset, the a spectrum is depicted together with the model, which includes a decays from ²³⁸U and ²³²Th families.

Energy spectrum of α particles registered by ZnWO₄ detector



$$\begin{split} T_{1/2} &= 10^9 \text{ yr } [1,2] \\ S &= 7 \text{ events (with 0 background, very conservatively)} \rightarrow \lim S < 11.77 \text{ at } 90\% \text{ C.L.} \\ \lim S &= \ln 2 \cdot \varepsilon \cdot N \cdot t \ / T_{1/2} \rightarrow N(\text{Sb}) \\ N(\text{Sg}) / N(\text{W}) < 5.5 \times 10^{-14} \text{ atoms/atom at } 90\% \text{ C.L.} \end{split}$$

[1] Rep. Prog. Phys. 47 (1983) 817
 [2] AIP Conf. Proc. 1175 (2009) 297

This value is comparable with the sensitivity reached in the searches for eka-Os in the SHIN experiment $< 10^{-14}$ g/g [2]

CdWO₄ crystals

- good scintillation properties
- "source = detector approach" (~100% efficiency)
- low levels of internal contamination
- particle discrimination ability (\ background)
- possibility to search SHE

CdWO₄ were successfully used in lowbackground experiments on search for 2 β decay of Cd and W [1], as well as for the study of rare α [2] and β [3] decays



J.D. Vergados et al., RPP 75(2012)106301

^{116}Cd - One of the most promising isotopes to search for $0\nu2\beta$ decay

- $Q_{2\beta} = 2813.44(13) \text{ keV}$
- $\delta = 7.5\%$
- promising theoretical calculation
- possible isotopic enrichment in large amount

ZPA 355(1996)433, EPJA 36(2008)167, PRC 93(2016)045502;
 PRC 67(2003)014310;
 PAN 59(1996)1, PRC 76(2007)064603

¹¹⁶CdWO₄ crystal scintillator



[1] JINST 6(2011)P08011

The optical transmission curve of ¹¹⁶CdWO₄ before and after annealing Attenuation length is 60 cm

Good optical and scintillation properties of the crystal were obtained thanks to the deep purification of ¹¹⁶Cd and W, and the advantage of the low-thermal-gradient Czochralski technique to grow the crystal [1]

Boule of enriched 116 CdWO₄ crystal (82% of 116 Cd). The conic part of the boule is the beginning of the crystal growth.

Yield of the crystal boule is 87% of the initial powder Losses (the total production cycle) < 3%



Experimental set-up with ¹¹⁶CdWO₄



Laboratori Nazionali del Gran Sasso, Italy (3600 m we)

Experimental set-up with ¹¹⁶CdWO₄



Pulse shape discrimination (PSD), 26831 h



Shape indicator (SI) versus energy for the background exposure (26831 h \times 1.162 kg)

The optimal filter method proposed by E. Gatti and F. De Martini, developed for CdWO scintillation detectors [1 and ref.]

[1] Nucl. Instrum. Methods Phys. A 410 (1998) 213 (1998)

$$P(t) = [f_{\alpha}(t) - f_{\gamma}(t)] / [f_{\alpha}(t) + f_{\gamma}(t)],$$

 $f_{\alpha}(t), f_{\gamma}(t)$ – shapes of the signals

Selection of ²¹²Bi-²¹²Po events by front-edge analysis



Sum energy spectrum of α events selected by PSD



The fit of the data by the model built from α decays of U and Th with daughters, and residual γ , β background

Time-amplitude analysis





M.J. Koskelo et al., Radioact. Radiochem. 7(1996)18

$$f(u) = \begin{cases} A \exp\left[-\frac{(u-\mu)^2}{2\sigma^2}\right], & \text{if } u \ge \mu - T\\ A \exp\left[\frac{T(2u-2\mu+T)}{2\sigma^2}\right], & \text{if } u < \mu - T \end{cases}$$

- A amplitude of Gauss
- μ center of Gauss
- σ standard deviation

T determines both the characteristics of the tailing and its joining point with the Gaussian

Activity ²²⁸Th, μ Bq/kg Alpha peaks of ²²⁴Ra, ²²⁰Rn and ²¹⁶Po selected by the time-amplitude **PSD** T-A analysis from the data accumulated during 26831 h with the Crystal 1 17(2) 17(1) 1. The obtained $^{116}CdWO_{4}$ detector No. half-lives of Crystal 2 26(2) 27(1) 220 Rn (58 ± 4 s) and 216 Po (0.136 ± 0.006 s) are 22 agreement with 22 + α/γ ratio the table values (55.6 \pm 0.1 s and 0.145 \pm 0.002 s, respectively [TOI]).

Dependence of α/γ ratio on energy of the α particles



 α/γ ratio = 0.114(7) + 0.0133(12) E_{α}

Radioactive contaminations of ¹¹⁶CdWO₄ crystal scintillators (and elements of the set-up)

Chain	Nuclide	Activity, mBq/kg
²³² Th	²³² Th	0.07(2)
	²²⁸ Th	0.020(1)
²³⁸ U	²³⁸ U	0.58(4)
	²³⁴ U	0.6(1)
	²³⁰ Th	≤ 0.13
	²²⁶ Ra	≤ 0.006
	²¹⁰ Pb	0.70(4)
	¹¹⁶ Cd	1.138(5)
	⁴⁰ K	0.22(9)
	^{110m} Ag	< 0.007

	Nuclide	Activity, mBq/kg
PMT	²²⁶ Ra	$<0.9 \times 10^{3}$
	²²⁸ Ra	$0.12(5) \times 10^{3}$
	²²⁸ Th	$0.83(2) \times 10^2$
	⁴⁰ K	<13×10 ³
Copper	U	0.11(2)
	Th	0.06(1)
	⁴⁰ K	0.27(6)
Light guides	U	0.20(2)
	Th	0.10(1)
	⁴⁰ K	1.3(3)

Total α activity of two crystals = 2.14(2) mBq/kg

Two neutrino double beta decay of ¹¹⁶Cd (25037 h)



Signal to bg ratio: 2.6 in [1.1–2.8] MeV

Index of Bg: 0.12 in [2.7-2.9] MeV

 $T_{1/2} = [2.69 \pm 0.02 (\text{stat.}) \pm 0.14 (\text{syst.})] \times 10^{19} \text{ yr}$

Summary of the $T_{1/2}(2\nu 2\beta)$ results ¹¹⁶Cd



J. Phys. Soc. Japan 64(1995)339; [2] Phys. Lett. B 344(1995)72;
 Z. Phys. C 72(1996)239; [4] PRC 62(2000)045501;
 PRC 68(2003)035501; [6] AIP Conf. Proc. 1572(2013)110;
 PRC 81(2010)035501; [8] NPA 935(2015)52;
 PRD 95(2017)012007.

Results

Decay mode	Transition, level of ¹¹⁶ Sn (keV)	$T_{1/2}$ (yr)
2ν	g.s.	$(2.63^{+0.11}_{-0.12}) \times 10^{19} \text{ yr}$
2ν	2+ (1294)	$\geq 9.8 \times 10^{20}$
2ν	0+ (1757)	$\geq 5.9 \times 10^{20}$
2ν	0+ (2027)	$\geq 1.1 \times 10^{21}$
2ν	2+ (2112)	$\geq 2.5 \times 10^{21}$
2ν	2+ (2225)	$\geq 7.5 \times 10^{21}$
0ν	g.s.	$\geq 2.2 \times 10^{23}$
0ν	2+ (1294)	$\geq 7.1 \times 10^{22}$
0ν	0+ (1757)	$\geq 4.5 \times 10^{22}$
0ν	0+ (2027)	$\geq 3.1 \times 10^{22}$
0ν	2+ (2112)	$\geq 3.7 \times 10^{22}$
0ν	2+ (2225)	$\geq 3.4 \times 10^{22}$
$0\nu\chi^0 n = 1$	g.s.	$\geq 8.2 \times 10^{21}$
$0\nu\chi^0 n = 2$	g.s.	$\geq 4.1 \times 10^{21}$
$0\nu\chi^0 n = 3$	g.s.	$\geq 2.6 \times 10^{21}$
$0\nu\chi^0\chi^0n=3$	g.s.	$\geq 2.6 \times 10^{21}$
$2\nu LVn = 4$	g.s.	$\geq 1.2 \times 10^{21}$
$0\nu\chi^0\chi^0 n=7$	g.s.	$\geq\!\!8.9\times10^{20}$

[1] PRC 68(2003)035501 [2] Phys.Lett.B 249(1990)186 *68% C.L. [3] NPA 577(1994)493

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First results

Counts / 20 keV $m(det1) = 579.8 \text{ g} \rightarrow N_1(W) =$ 26831 h 9.67×10^{23} nuclei 10^{2} $m(det2) = 582.4 \text{ g} \rightarrow N_2(W) =$ 9.71×10^{23} nuclei 8.9 MeV 12.5 MeV $T_{1/2}$ (Sg) = 10⁹ yr [1,2] – standard 10 assumption in the SHE search in nature. 1 $\lim S_1 < 193$ $\lim S_2 < 199$ 1000 1500 2000 2500 3000 3500 Energy, keV

> $N(Sg)_1/N(W) \le 9.8 \times 10^{-14}$ atoms/atom at 90% C.L. $N(Sg)_2/N(W) \le 1.0 \times 10^{-13}$ atoms/atom at 90% C.L.

[1] Rep. Prog. Phys. 47 (1983) 817
 [2] AIP Conf. Proc. 1175 (2009) 297

This values are comparable with the sensitivity reached in the searches for eka-W with $ZnWO_4 < 5.5 \times 10^{-14} \text{ g/g [2]}$

Search for eka-B [Cardani et al., JINST 7(2012)P10022]



LNGS, BGO scintillating bolometer (891 g), t = 455 h 3 α events in the energy interval 9.5–10 MeV (while one could expect no events after the energy of the most energetic in the U/Th chains α particles from Po with Q = 8.954 MeV). If they are not pile-ups of two α

signals or BiPo event, then \rightarrow

If S < 6.68 at 90% C.L., $T_{1/2}$ (Sb) = 10⁹ yr for eka-Bi N(eka-Bi) =1.9 × 10¹¹. N(Bi) =1.7 × 10²⁴

 $N(eka-Bi)/N(Bi) < 1.1*10^{-13} atoms/atom$

This is better than the limits obtained in recent searches with the accelerator mass spectrometry $\delta < (5-30) \times 10^{-13}$ atoms/atom (for A= 293-300) demonstrating the good potentiality of the approach considered here to search for SHE

Thank you for attention!