Search for long-lived superheavy eka-tungsten with radiopure ZnWO₄ crystal scintillator

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Abstract
The data collected with a radioactively pure ZnWO₄ crystal scintillator (699 g) in low background measurements during 2130 h at the underground (3600 m w.e.) Laboratori Nazionali del Gran Sasso (INFN, Italy) were used to set a limit on possible concentration of superheavy eka-W (seaborgium Sg, Z = 106) in the crystal. Assuming that one of the daughters in a chain of decays of the initial Sg nucleus decays with emission of high energy α particle (Qα > 8 MeV) and analyzing the high energy part of the measured α spectrum, the limit N(Sg)/N(W) < 5.5 × 10¹⁴ atoms/atom at 90% C.L. was obtained (for Sg half-life of 10⁹ yr). In addition, a limit on the concentration of eka-Bi was set by analysing the data collected with a large BGO scintillation bolometer in an experiment performed by another group (Cardani et al 2012 JINST 7 P10022): N(eka-Bi)/N(Bi) < 1.1 × 10¹³ atoms/atom with 90% C.L. Both the limits are comparable with those obtained in recent experiments which instead look for spontaneous fission of superheavy elements or use the accelerator mass spectrometry.

Keywords: superheavy elements, ZnWO₄ crystal scintillator, low background experiment

(Some figures may appear in colour only in the online journal)

1. Introduction
Possible existence of superheavy elements (SHEs) with atomic masses A ≥ 250 and atomic numbers Z ≥ 104 was already discussed in the 1950s [1]. In the 1960s, the development of new methods of calculation of the shell model corrections to the liquid drop model predicted a neutron-rich ‘island of stability’ around the double magic Z = 114 or 126, N = 184 [2–8], with half-life of the nucleus ²⁶⁸Db calculated as 10⁵ yr [7] and 2.5 × 10⁹ yr [8]. Various recent calculations [9–16] related to different macro–micro and microscopic models predict N = 184 as the magic number of neutrons and Z = 114, 120 or 126 as the proton magic number for spherical nuclei.

In experiments on the artificial synthesis of the SHE in fusion of ions with accelerators, more than one hundred different unstable isotopes with Z = 104 − 118 were created [17–20], with half-lives from microseconds to hours (for ²⁶⁸Db T½ = 29.4 s [18]). New isotopes with Z = 107 − 118 predominantly undergo a chain of α decays followed by spontaneous fission (SF) [19]. Note that the SHEs formed in fusion reaction [17, 18] are proton-rich (or neutron-deficient), and the half-lives of SHE with number of neutrons near the magic number 184 are expected to be longer.
While maybe 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 and $\beta^-$ decays [21] (for current status of our understanding of SHE nucleosynthesis see e.g. [22–24] and references therein). It should be noted that some recent estimations (e.g. [23, 25]) are more pessimistic: calculations [23] predict that superheavy nuclei with masses up to $A \approx 300$ can be produced during star explosions but they decay through several $\beta$ transformations and spontaneous fission on time scales of days not reaching the valley of stability. Such results, however, evidently depend on models (and many theoretical parameters) used for description of stars, their evolution and nucleosynthesis during explosion, as well as on models which predict masses of nuclei and their half-lives, respectively, to $\alpha$, $\beta$ decays and spontaneous fission. Thus, the possibility that long-lived SHEs were synthesized in the r-process and that they are still present in minor quantities in Earth materials is not fully excluded.

In the 1970s and 1980s, an extensive program to search for SHEs in nature was undertaken. Hundreds of samples from the ocean floor to the lunar surface were analyzed with sensitivity to mass concentration of $10^{-11} – 10^{-12}$ g/g (see reviews [26–30] and references therein). Some hints were obtained on the presence of SHEs through their fission in meteorites and in hot spring waters from the Cheleken peninsula (Caspian sea), or through their long tracks in olivine crystals from meteorites (which supposedly belong to SHEs with $Z \approx 110$). Giant radioactive halos in minerals were also considered as possibly caused by radiation damage due to $\alpha$ particles with energy 10–15 MeV emitted from the central inclusion by SHEs with $Z \approx 120$ [31, 32]. While the old results are summarized in [26–30], we will give details below on more recent investigations.

In the measurements of Marinov et al with natural gold using inductively coupled plasma sector field mass spectrometry, superheavy isotopes with atomic masses $A = 261$ and 265 and concentration $\delta = (1–10) \times 10^{-10}$ atoms/atom relatively to Au were found [33]. It was proposed that they are most probably isotopes of roentgenium Rg (eka-Au, $Z = 111$). In similar studies on natural thorium, superheavy nuclei with $A = 292$ and $\delta \approx 10^{-12}$ atoms/atom relatively to $^{232}$Th were found, interpreted as possible existence of long-lived eka-Th ($Z = 122$) [34]. It should be noted, however, that the evidence on the existence of SHEs with $A = 261, 265$ in natural Au was not confirmed in sensitive searches with accelerator mass spectrometry (AMS) of the Vienna group [35] where few orders of magnitude lower limits of $\delta < 3 \times 10^{-16}$ atoms/atom were obtained. Similarly, only a limit of $\delta < 4 \times 10^{-15}$ atoms/atom was found for abundance of SHEs with $A = 292$ in Th [36] (see also comments in [37]). The AMS searches of the same group for SHEs in natural Pt, Pb and Bi also gave only the limits [38]: (i) $\delta < (0.9–2) \times 10^{-15}$ atoms/atom for eka-Pt (Ds, $Z = 110$) in Pt for SHE atomic masses $A = 288 – 295$; (ii) $\delta < (2 – 6) \times 10^{-14}$ atoms/atom for eka-Pb (Fl, $Z = 114$) in Pb (for $A = 292, 293, 295 – 297, 299$); (iii) $\delta < (5 – 30) \times 10^{-13}$ atoms/atom for eka-Bi ($Z = 115$) in Bi (for $A = 293 – 300$).

Recently the AMS technique was also used in the searches of the Garching group for SHEs with $292 \leq A \leq 310$ in samples of Os, Pt and PbF$_2$, where only limits were established in the range of $1.5 \times 10^{-16}–4.1 \times 10^{-14}$ atoms/atom [39].

In the OLIMPIYA experiment [40], in the analysis of $\approx 6000$ tracks from cosmic rays accumulated during $(1 – 3) \times 10^8$ yr in $\approx 170$ olivine crystals extracted from meteorites, three tracks were found which could belong to nuclei with $105 < Z < 130$. A more accurate estimation for one of them gave $Z = 119_{-6}^{+7}$ at 95% C.L. [40, 41].

Analyzing recent theoretical calculations, Oganessian concluded [18, 42] that the most stable superheavy nuclide could not be eka-Pb with $Z = 114$ but nuclides with $A \approx 290$ and $Z = 106, 107, 108$ (eka-W, eka-Re, eka-Os, respectively). It was expected that they will decay through a chain of $\beta$ and $\alpha$ decays which may end with spontaneous fission. It was proposed [42] to use thin foils ($\approx 0.1$ mg cm$^{-2}$) to register expected $\alpha$ particles (this immediately constrains masses of samples on the scale of grams).

A search for eka-Os (Hs, $Z = 108$) was carried out in the experiment SHIN (SuperHeavy In Nature) deep underground (4800 m.w.e., to suppress background from neutrons created by cosmic muons) in the Laboratoire Souterrain de Modane (France) [43]. It was expected that either the initial Hs nucleus or its daughter created in a chain of $\alpha$ decays will finally decay through SF. The average number of neutrons per fission is equal to $\bar{\nu} \approx 4.5$ for $Z = 104$ and $\bar{\nu} \approx 6$ for $Z = 108$, very different from that from possible background from SF of $^{235}$U ($\bar{\nu} \approx 2$). An Os sample with mass of 550 g was measured during 3 yr with a neutron detector which consisted of 60 $^3$He counters placed in 4 rings around the sample. Few events with multiplicity $\nu \geq 3$ were observed, but only a limit on mass concentration of eka-Os in Os $\delta \leq 10^{-14}$ g/g was set (with the standard assumption that the half-life of eka-Os is $T_{1/2} = 10^9$ yr). Afterwards the Os sample was changed with a Xe sample (140 g) to search for a superheavy homolog of Xe; the sensitivity was estimated as $\delta \approx 10^{-13}$ g/g.

In the present work, we use an alternative approach: instead of searching for the spontaneous fission, we look for high energy $\alpha$ particles ($Q_{\alpha, \gamma} > 8$ MeV) possibly emitted in a chain of decays of eka-W (seaborgium Sg, $Z = 106$) and registered by a radioactively pure ZnWO$_4$ crystal scintillator working in a low background installation at the Laboratori Nazionali del Gran Sasso (LNGS) of the INFN (Italy) at a depth of 3600 m.w.e. Chemical properties of seaborgium are similar to those of tungsten [44–46], and it is expected that Sg in some amount follows W in processes of chemical purification and growth of a ZnWO$_4$ crystal$^7$. Such a technique, when a source of radiation is embedded in a detector

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$^7$ Because of the actinoid contraction, the atomic and ionic radii of trans-actinoids in the 7th period are expected to be close to the radii of their homologs in the previous period of the periodic table, in the same way as the lanthanoid contraction makes the radii of pre- and post-lanthanoid homologs similar. Thus, one should expect that the trans-actinoids would easily substitute their homologs in crystal lattices.
contours were published [49]. Here we reanalyze the data for possible presence of SHEs in ZnWO4 crystals.

2. Experiment and data processing

The detailed description of the set-up with ZnWO4 crystal scintillators and its performance have been discussed in [47–49]. Four ZnWO4 detectors were measured (with mass of 117, 141, 239 and 699 g). While their characteristics were very similar, the largest of them was also found to be the most radioactively pure [49], and in the following we will consider only that in more detail. Here we recall the main features of the measurements.

The ZnWO4 crystal scintillator (44 × 55 mm, mass of 699 g) was grown by the Czochralski method. It was fixed inside a cavity of 47 × 59 mm in the central part of a polystyrene light-guide 66 mm in diameter and 312 mm in length. The cavity was filled up with high purity silicone oil. The light-guide was optically connected on opposite sides by optical couplant to two low radioactive light-guide EMI:9265–B53/FL 3° photomultipliers (PMT). The light-guide was wrapped by PTFE tape.

The detector has been installed deep underground (∼3600 m w.e.) in the low background DAMA/R&D set-up at the LNGS. It was surrounded by Cu bricks and sealed in a low radioactive airtight Cu box continuously flushed with high purity nitrogen gas to avoid the presence of residual environmental radon. The copper box was surrounded by a passive shield made of 10 cm of high purity Cu, 15 cm of low radioactive lead, 1.5 mm of cadmium and 4/10 cm polyethylene/paraffin to reduce the external background. The whole shield has been closed inside a plexiglas box, also continuously flushed by high purity nitrogen gas.

An event-by-event data acquisition system accumulates the amplitude and the arrival time of the events. The profile of the sum of the signals from the PMTs was also recorded with a sampling frequency of 20 MS s⁻¹ over a time window of 100 μs by an 8 bit transient digitizer (DC270 Acqiris). A CAMAC system was used to manage the triggers and the ADCs, introducing a rather long dead time of about 26 ms (in particular, because of storing the time profiles on a hard disk).

The energy scale and energy resolution of the ZnWO4 detector have been measured with 22Na, 133Ba, 137Cs, 228Th and 241Am γ sources. The energy resolution of the ZnWO4 detector for γ quanta is well described as FWHM(keV) = \sqrt{13.96(47) \times E_γ}, where \( E_γ \) is the energy of γ quanta in keV.\(^8\)

\(^8\) Because γ rays interact with matter by means of the energy transfer to electrons, the same is applicable also for β particles; we use abbreviation ‘γ(β)’ in such cases in the following.

It is known that light yield for α particles in scintillators is lower than that for γ quanta (β particles) of the same energy [50]. The ratio of the α peak position in the energy scale measured with γ sources to the real energy of the α particles for \( E_α > 2 \) MeV is described as \( \alpha/\beta = 0.074(16) + 0.0164(40) \times E_α \), where \( E_α \) is the energy of α particles in MeV. The energy resolution for α particles is: FWHM, (keV) = 33 + 0.247 × \( E_α^γ \), where \( E_α^γ \) is energy of α particles in γ scale (in keV).\(^9\)

The scintillation signals induced in ZnWO4 by α particles are different from those caused by γ(β) particles (signals from α’s are shorter) [51]. This difference allows one to discriminate γ(β) events from α events. We used for this purpose the optimal filter method [52]. For each signal, the numerical characteristic of its shape (shape indicator, SI) was defined as \( SI = \sum f(t_k) \times P(t_k) \sum f(t_k) \), where the sum is over the time channels \( k \), starting from the origin of signal and up to 50 μs; \( f(t_k) \) is the digitized amplitude (at the time \( t_k \)) of a given signal. The weight function \( P(t) \) was defined as: \( P(t) = [f_\alpha(t) - f_\beta(t)]/[f_\alpha(t) + f_\beta(t)] \), where \( f_\alpha(t) \) and \( f_\beta(t) \) are the reference pulse shapes for α particles and γ quanta, respectively, obtained by summing up the shapes of a few thousand γ or α events. The scatter plot of the shape indicator versus energy for the data of the low background measurements with the ZnWO4 detector during 2130 h is shown in figure 1. The distributions of the SI values for the α and γ(β) events are well described by Gaussian functions, with center and width dependent on energy (the SI distributions for events in the energy interval 800–900 keV are shown in the inset of figure 1). Using these dependences, ±2σ contours were calculated where 95% of the corresponding events are contained. Detailed analysis of the obtained γ(β) and α spectra is

\(^9\) The energy resolution for α particles is worse than that for γ quanta due to the dependence of the \( \alpha/\beta \) ratio on the direction of the α’s relative to the ZnWO4 crystal axes [51].
given in [47]; clusters within the γ(β) limits belong to ^{40}K, ^{66}Zn, ^{208}Tl, ^{214}Bi, and alpha events are related mainly with ^{238}U and its daughters. More details on measurements and data processing can be found in [47–49].

A one-dimensional energy spectrum of α events inside the ±2σ contour is shown in figure 2 where the real energies of the α particles were obtained using the α/β ratio given above.

3. Limit on long-lived Sg in ZnWO₄ crystal

As was shown in [44–46], the chemical properties of superheavy Sg are similar to those of W; thus, one could expect that long-lived Sg follows W in the processes of chemical separation and growth of the ZnWO₄ crystals, and could also be present at some amount in the ZnWO₄ detector. If in a chain of decays of initial Sg nucleus an α decay with high Qα value occurs, we can see it in the high energy part of the spectrum presented in figure 2.

The superheavy isotopes can decay through emission of β– particle or by electron capture EC (or β+ decay), α decay or spontaneous fission; cluster decay also starts to be important at higher Z values (see recent reviews [15, 53]). In general, energy release Qα in α decay is increasing with the increase of the atomic number Z; in accordance with [54], experimental Qα values for superheavy elements with Z = 102 – 107 lie in the interval 7.8–10.6 MeV.

The decay of superheavy Sg (Z=106) will lead to independent events in the high energy part of the α spectrum in the following scenario:

1. Long-lived initial Sg nucleus decays through β– channel (thus creating a nucleus with Z=107) or through EC/β+ (leading to a nucleus with Z=105) or through α decay (resulting in Z=104). The energy of this first α decay should be quite modest (≈4 – 6 MeV, similar to that in decay of ²³²Th and ²³⁵,²³⁸U) to be consistent with the large T¹/₂. Such a low energy, however, does not allow one to distinguish it from the decays in usual U/Th chains present in trace amount in any material;

2. The created nucleus (or one of its daughters) decays with emission of high energy α particle (Qα > 8 MeV). The energy threshold of 8 MeV allows one to cut off contributions from α decays in U/Th chains because all nuclides in these chains have Qα < 8 MeV, with the exception of ²¹²Po with Qα = 8.954 MeV [55];

3. This daughter nuclide with Qα > 8 MeV, nevertheless, should live long enough to be registered outside of the dead time of the data acquisition system after the preceding decay (in our measurements it is 26 ms¹⁰). This is also realistic because many of the experimentally discovered to-date SHE isotopes with Qα > 8 MeV have T¹/₂ in the range of seconds (or larger) [19, 54]. The theoretical predictions [56–58] also confirm this assumption.

It is difficult to recognize exactly initial Sg nuclide and its chain of decays which fulfill all the above conditions. One of the preconditions could be the α decay with half-lives relative to spontaneous fission and α decay estimated in [9] as: T¹/₂ = 1.3 × 10⁶ yr and T¹/₂ = 1.6 × 10¹¹ yr. Only limit for β decay was given in [10]: T¹/₂ > 100 s, and this nucleus is quite stable with respect to cluster radioactivity: T¹/₂ ≃ 10²⁰ yr [59]. However, the expected T¹/₂ values for all possible decay channels of all potential daughters of ²⁹⁰Sg (and its neighbours) are not available in the literature. One has also to remember that all theoretical estimations strongly depend on the models (and their parameters) used for calculation of nuclear masses and half-lives for α, β/EC, SF and cluster decay channels (just for example, in the above mentioned works T¹/₂ of ²⁹⁰Sg was estimated as: 1.6 × 10¹¹ yr [9], 9.1 × 10⁶ yr [10]). Taking into account such theoretical uncertainties, experimental investigations should be performed too.

If conditions (1)–(3) are fulfilled, we can use the data collected in the low background measurements with the ZnWO₄ scintillator during t = 2130 h to restrict the presence of long-lived Sg in the crystal. As one can see in figure 2, we have S = 7 events in the energy interval 8–15 MeV. Some of them (at lower energies) could be related with ²¹²Po, some of them (at higher energies) could be γ(β) events (see figure 1) or the so-called ²¹²Bi-²¹²Po events [48], but here we conservatively take all of them to derive the limit on number N of Sg nuclei. For time of measurements t < T¹/₂ one can use relation: S = ln 2 · ε · N · t/T¹/₂, where ε is the efficiency to register α decay in the ZnWO₄ detector. It is clear that the probability to absorb α particle in the large (699 g) crystal is practically equal to 1. We cannot estimate loss of efficiency due to the dead time of the data acquisition because half-life of α decaying nucleus is unknown. Thus, in the following we will use only ε = 0.95 determined by the cuts in the selection

¹⁰ It should be noted that very fast chains of decays can also be found: by analysis of the time profile of events, which is recorded in our measurements during 100 μs. However, this is outside the present work.
of the $\alpha$ events (see figure 1). For the half-life of the initial Sg nucleus, we will adopt the value: $T_{\alpha/2} = 10^9$ yr which is a standard assumption in the SHE searches in nature [28, 43]. With the measured number of $S = 7$ events and very conservatively supposing 0 background, the number of decays is limited by $\Sigma \leq 17.77$ at 90% C.L. in accordance with [60]. With all the values given above, one obtains the limit on the number of Sg nuclei as: $\Sigma(N(Sg)) < 7.4 \times 10^{10}$. Since the number of W nuclei in the 699 g ZnWO$_4$ crystal is $N(W) = 1.34 \times 10^{24}$, one gets the concentration of Sg in ZnWO$_4$ relatively to W:

$$N(Sg)/N(W) < 5.5 \times 10^{-14} \text{ atoms/atom at 90}% \text{ C. L. \ (1)}$$

This value is comparable with the sensitivity reached in the searches for eka-Os in the SHIN experiment ($\delta < 10^{-14} \text{ g/g}$) [43].

However, the above limit has a drawback: it is given relatively not to natural W but relatively to W in our ZnWO$_4$, i.e., to W which passed processes of extraction from initial W-containing ores (commercially important minerals are mainly scheelite CaWO$_4$ and wolframite (Fe,Mn)WO$_4$), purification and growth of the ZnWO$_4$ crystal. It is difficult to estimate possible losses of superheavy Sg in all these processes but one can try to do this indirectly, using information on the lighter homolog of W: molybdenum. The following typical values can be found for content of Mo in W supplied by different companies: 150 ppm for 99.95% W (GoodFellow [61]), 30 ppm for 99.97% WO$_3$ and up to 3000 ppm for W (Wolfram [62]), 100 ppm for 99.998% (except Mo) WO$_3$ (Alfa Aesar [63]), 12 ppm for 99.97% W (PLANSEE [64]). If we conservatively assume 10 ppm content of Mo in WO$_3$ powder used in production of the ZnWO$_4$ crystal and 1% content of Mo in an initial W-containing mineral (both these values are unknown for our ZnWO$_4$), we can estimate a conservative limit on the presence of eka-Bi in the BGO. In accordance with the procedure given above one gets the limit on the number of events: $S < 6.68$ at 90% C.L. [60]. Supposing once more $T_{\alpha/2} = 10^9$ yr for eka-Bi, this number of decays during 455 h corresponds to a number of eka-Bi nuclei as $N(eka-Bi)=1.9 \times 10^{11}$. The number of the Bi nuclei in the 891 g BGO is: $N(Bi)=1.7 \times 10^{24}$, and thus the limit on the eka-Bi concentration in BGO is $\delta < 1.1\cdot 10^{-13}$ atoms/atom relatively to Bi. This is better than the limits obtained in recent searches with the accelerator mass spectrometry $\delta < (5-30) \times 10^{-13}$ atoms/atom (for $A = 293 - 300$) [38], demonstrating the good potentiality of the approach considered here to search for SHE. It should be remembered, however, that this limit is also obtained relatively not to natural Bi but for Bi in the BGO crystal after processes of Bi extraction, purification and the BGO crystal growth. While one could expect a reduction factor for natural Bi similar to that in case of W, this question has to be studied additionally.

5. Conclusions

The data collected with a radioactively pure ZnWO$_4$ crystal scintillator in low background measurements during 2130 h at LNGS were used to set a limit on possible concentration of superheavy eka-W (seaborgium Sg, $Z = 106$) in the crystal. Assuming that one of the daughters in a chain of decays of the initial Sg nucleus decays with emission of high energy $\alpha$ particles ($Q_\alpha > 8$ MeV) and that half-life of the long-lived Sg is $10^9$ yr (that is standard assumption in this field [28, 43]), we obtained the limit on Sg concentration as:

$$N(Sg)/N(W) < 5.5 \times 10^{-14} \text{ atoms/atom at 90}% \text{ C.L. \ (2)}$$

This is comparable with limits obtained with the accelerator mass spectrometry $\delta < (5-30) \times 10^{-13}$ atoms/atom for eka-Os in Os in the recent SHIN experiment [43]. One should note that the detection of spontaneous fission, as in SHIN, and the detection of high energy $\alpha$ particles, as here, are complementary approaches in the searches for SHE in nature.

The limit on the concentration of eka-Bi in Bi: $N(eka-Bi)/N(Bi) < 1.1 \times 10^{-13}$ atoms/atom was also set from the measurements with a large BGO scintillation bolometer of [66]. This is comparable with limits obtained with the accelerator mass spectrometry $\delta < (5-30) \times 10^{-13}$ atoms/atom (for $A = 293 - 300$) [38].

Both the limits for eka-W and eka-Bi were obtained relatively not to natural W or Bi but for W and Bi present in ZnWO$_4$ and BGO crystals, i.e., after processes of extraction,
purification and crystals growth. Recalculated for natural W and Bi, limits can be lower by a factor of $\sim 10^3$.

The approach followed in this work can be also used in searches for other superheavy elements as eka-Tl in NaI(Tl), eka-Pb in PbWO$_4$, and superheavy homolog of Xe in xenon. It should be noted that detection of $\alpha$ particles with $Q_\alpha > 8$ MeV cannot definitively point to a specific isotope (e.g. eka-W or eka-Bi). However, if events with shape of scintillation signal typical for $\alpha$’s and high energy will be persistently registered in convincing amounts and alternative explanations (like Bi-Po events, pile-ups) will be absent, this would be an indication on presence of SHE and necessity to involve also other methods for investigation.

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