Search for naturally occurring seaborgium with radiopure $^{116}CdWO_4$ crystal scintillators

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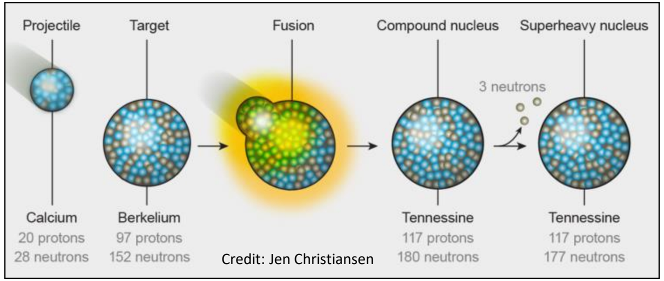
Long-lived superheavy elements (SHEs)

Hypothesis on the existence of long-lived SHEs (A>250, Z>104) was discussed since the 1950s

Half-lives of 10^8–10^9 yr were predicted for some nuclei in an island of stability around the double magic numbers Z=114 or 126, N=184 [1,2,3]

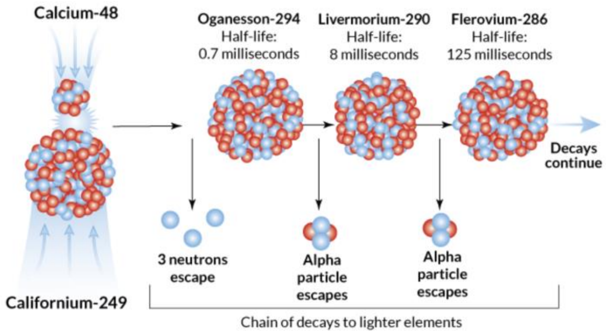
Artificial syntheses of SHEs with Z=104…118 were performed in fusion of heavy ions in accelerators [4]:

- The produced SHEs are neutron-deficient and short-living (up to ≈30 h)
- They don’t reach the expected filled neutron shell of N=184 which would stabilize the nuclei



Credit: Jen Christiansen

But, long-lived SHE could be produced in the natural conditions of intensive neutron flux in energetic stellar events like neutron star merging or supernovae explosions



T. TIBBITTS

It is expected that they would decay through a chain of α and β decays which ends with a spontaneous fission

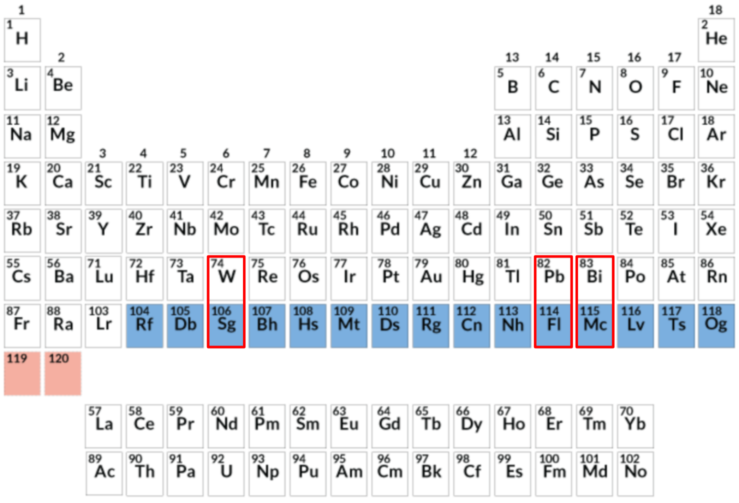
[1] SG Nilsson *et al* 1968 *Nucl. Phys. A* **115** 545
[2] SG Nilsson *et al* 1969 *Nucl. Phys.* A **131** 1
[3] EO Fiset and JR Nix 1972 *Nucl. Phys. A* **193** 647
[4] SA Giuliani *et al* 2019 *Rev. Mod. Phys.* **91** 011001

Search for SHEs in nature

No definitive presence of long-lived SHEs has been detected so far

Some of the used techniques:

- Search for long tracks in old minerals, due to high E α particles, not emitted by usual natural α decays [1,2]
- Analysis with mass-spectrometric methods as ICP-MS and AMS [3]
- Measure of neutrons emission in SF; 2–3 times more neutrons are expected for SHEs (SHIN experiment [4])
- Search for tracks produced by SHEs (assuming their presence in cosmic rays) in olivine crystals in meteorites (OLIMPIYA exp. [5])
- Search for high E α particles in inorganic crystal detectors that contain chemical elements which can be carriers of possible SHEs (real-time, source=detector approach) [6,7]:
  - $ZnWO_4$, $CaWO_4$, and $CdWO_4$ for seaborgium (eka-W, Z=106)
  - $Bi_4Ge_3O_{12}$ (BGO) for moscovium (eka-Bi, Z=115)
  - $PbWO_4$ for flerovium (eka-Pb, Z=114) and seaborgium


E. OTWELL

[1] GN Flerov and GM Ter-Akopian 1983 *Rep. Prog. Phys.* **46** 817
[2] GM Ter-Akopian and SN Dmitriev 2015 *Nucl. Phys. A* **944** 177
[3] P Ludwig *et al* 2012 *Phys. Rev. C* **85** 024315
[4] A Svirikhin A *et al* 2009 *AIP Conf. Proc.* **1175** 297
[5] V Alexeev *et al* 2016 *Astrophys. J.* **829** 120
[6] P Belli *et al* 2015 *Phys. Scr.* **90** 085301
[7] P Belli *et al* 2022 *Phys. Scr.* **97** 085302

Search for Sg with radiopure $CdWO_4$ crystal scintillators

[P Belli *et al* 2022 *Phys. Scr.* **97** 085302]

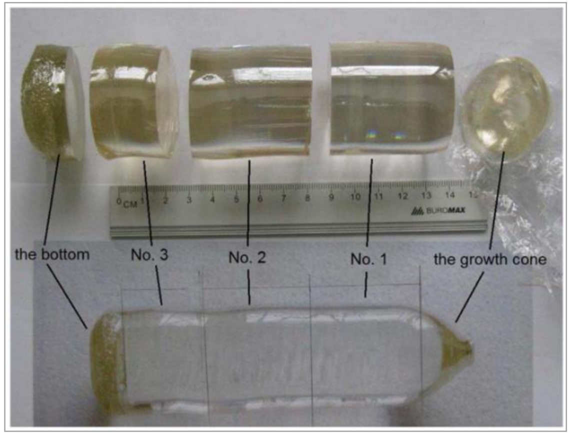
There exist predictions that Sg (Z=106) may be one of the most stable superheavy nuclides with bohrium (Z=107) or hassium (Z=108) [1]

The chemical properties of Sg are expected to be like those of its lighter homolog W:

- ○ the behavior of Sg is typical for a group 6 element of the periodic table, as W [2,3,4]
- ○ the calculated ionic radius of Sg^{6+} is 63pm [5], very close to that of W^{6+} (60pm)

⇒ Sg can replace W in crystals and follow tungsten in the processes of chemical separation and crystals growth of $CdWO_4$ crystal scintillators

⇒ The carrier of natural Sg is W in the $^{116}CdWO_4$ crystal scintillators



The use of $CdWO_4$ inorganic crystal detectors has some advantages such as:

- can be used as both a source and a detector
- The detection efficiency for the α events is almost 100%
- high particle discrimination ability to select the expected α events
- low level of internal radioactivity

⇒ Experimental signature: high energy α particles from the decay of a short-lived daughter nuclide of long-lived Sg

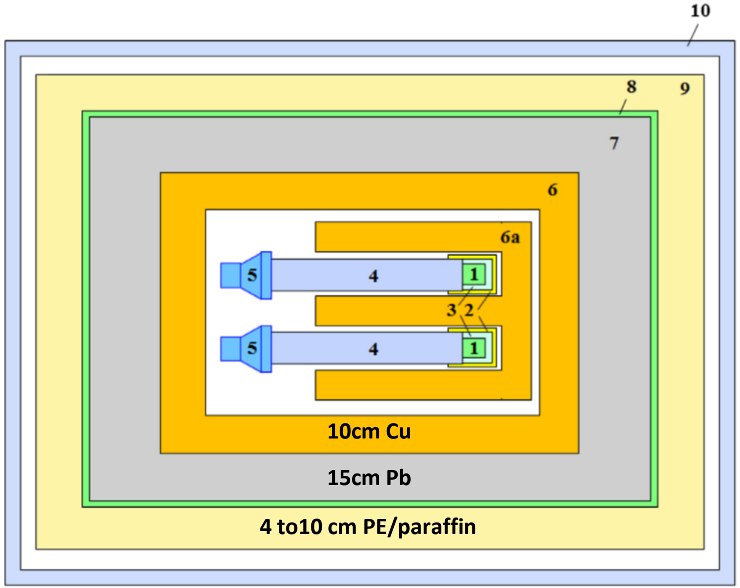
[1] Y Oganessian 2007 *J. Phys. G* **34** R165
[2] M Schädel *et al* 1997 *Nature* **388** 55
[3] V Pershina and J Anton 2013 *J. Chem. Phys.* **138** 174301
[4] J Even *et al* 2014 *Science* **345** 1491
[5] A Bilewicz 2000 *Radiochim. Acta* **88** 833

The experiment

[P Belli *et al* 2022 *Phys. Scr.* **97** 085302]

Reanalysis of the data of the **Aurora experiment** [1] (its main aim was the study of double beta decay of ^{116}Cd)

⇒ carried out in the low-background DAMA/R&D setup in the Gran Sasso Underground Laboratory (INFN, Italy)



Main setup components

1. Two ∅45×46 mm ^{116}CdWO$_4$ crystal scintillators (580g and 582g, Cd enriched in ^{116}Cd to 82%)
2. Teflon containers filled with
3. liquid scintillator
4. quartz light guides ∅7×40 cm
5. 3'' photomultipliers Hamamatsu R6233MOD
6. high-purity copper
7. low radioactive lead
8. cadmium
9. polyethylene/paraffin
10. Plexiglas box

Internal Cu box and external Plexiglas box flushed by purified N_2 gas to remove atmospheric radon

Pulse profile of each event recorded over 50 µs, with 20 ns time bin

[1] AS Barabash *et al* 2018 *Phys. Rev. D* **98** 092007

Data analysis: pulse shape discrimination (PSD)

[P Belli *et al* 2022 *Phys. Scr.* **97** 085302]

Optimal filter applied to discriminate γ(β) and α particles

Shape indicator (SI) calculated for each signal *f(t)*:

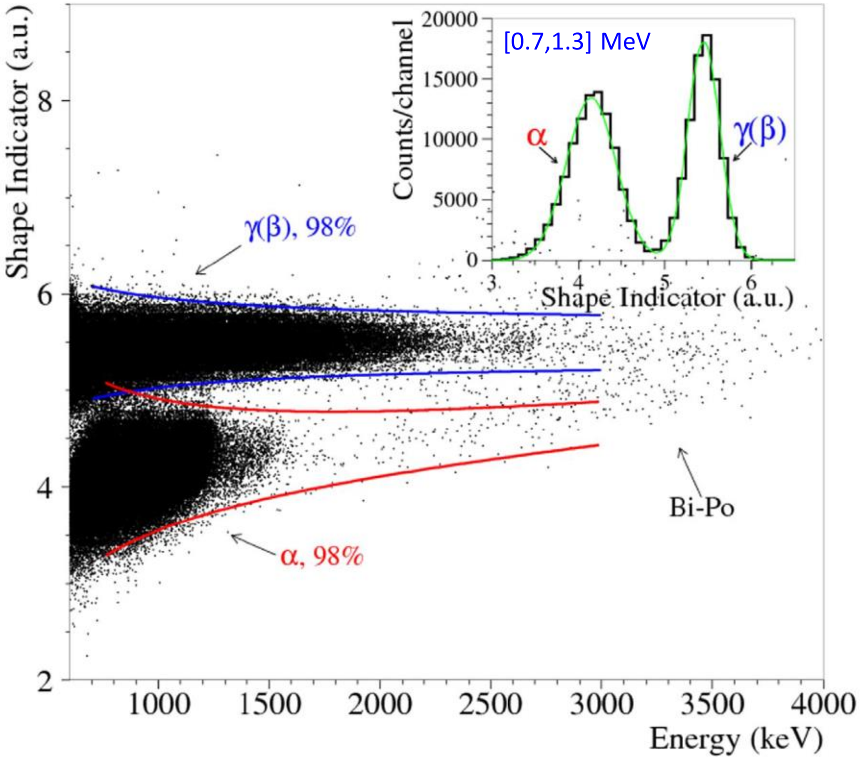
$$SI = \frac{\sum f(t_k)P(t_k)}{\sum f(t_k)}$$

$f(t_k)$ is the digital amplitude of a given signal at time t_k

$P(t) = \frac{f_\alpha(t)-f_\gamma(t)}{f_\alpha(t)+f_\gamma(t)}$ is the weight function

$f_\alpha(t)$ and $f_\gamma(t)$ are the reference pulse-shapes for α and γ(β) particles

From ^{116}CdWO$_4$ #2, T= 26831 h



Data analysis: front-edge analysis

[P Belli *et al* 2022 *Phys. Scr.* **97** 085302]

The front-edge analysis allows the identification of the Bi-Po events from the ^{232}Th decay chain:

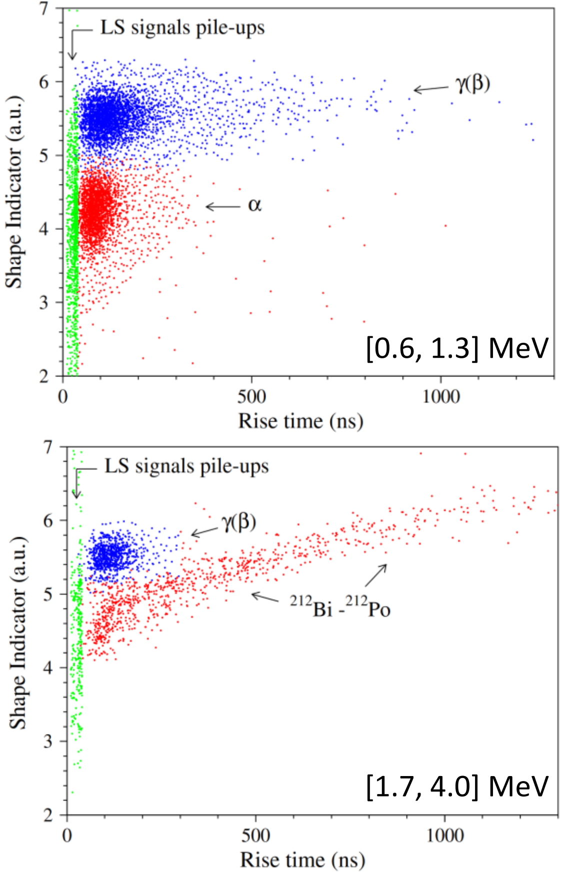
$^{212}Bi\ (Q_{\beta}=2252\ keV;\ T_{1/2}=60.55m)$

$\rightarrow {}^{212}Po\ (Q_{\alpha}=8954\ keV;\ T_{1/2}=295.1ns) \rightarrow {}^{208}Pb$

Rise time: calculated as time between the signal origin and the time where signal reach 0.7 of its maximal value

S.I. vs Rise-time

^{116}CdWO4 detector #2, T=26831 h



The identified Bi-Po events allowed to estimate the ^{228}Th activity:

- 0.018(2) mBq/kg for crystal #1
- 0.027(3) mBq/kg for crystal #2

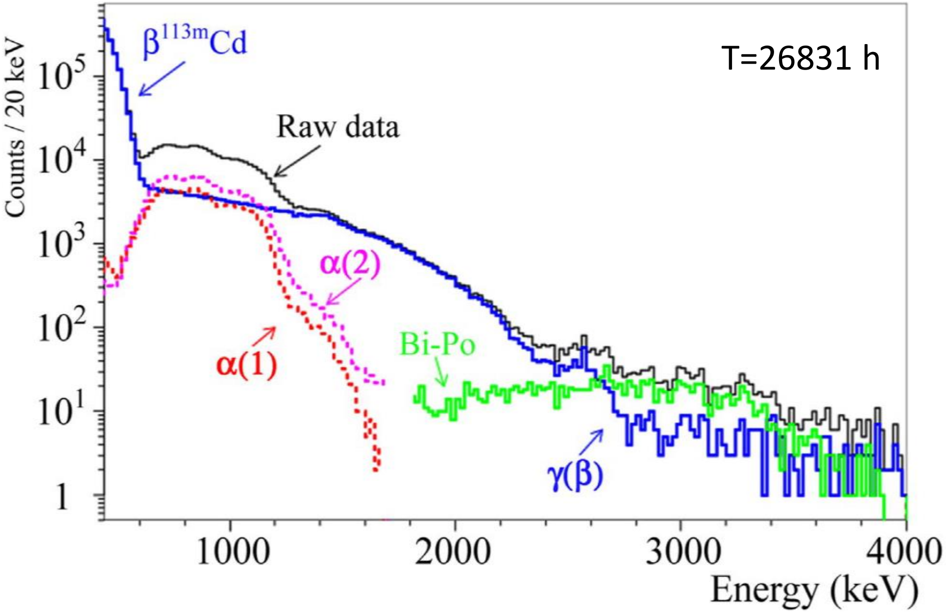
This analysis also allows the identification of pile-ups with signals produced by the liquid scintillator, thanks to the shorter rise time of its pulses (green dots)

Data analysis: total, α and $\gamma(\beta)$ spectra

[P Belli *et al* 2022 *Phys. Scr.* **97** 085302]

Energy spectra collected with the two $^{116}CdWO_4$ scintillators for:

- raw data (black)
- $\gamma(\beta)$ events selected by PSD analysis (blue)
- α events in crystal #1 and #2 selected by PSD analysis
- ^{212}Bi–^{212}Po events (Bi-Po) selected by PSD and front-edge analyzes



The efficiency of the pulse-shape selection for α events in the $^{116}CdWO_4$ is 96%

Total α activity is lower in crystal #1 due to segregation of the impurities in the crystal growth process [1]:

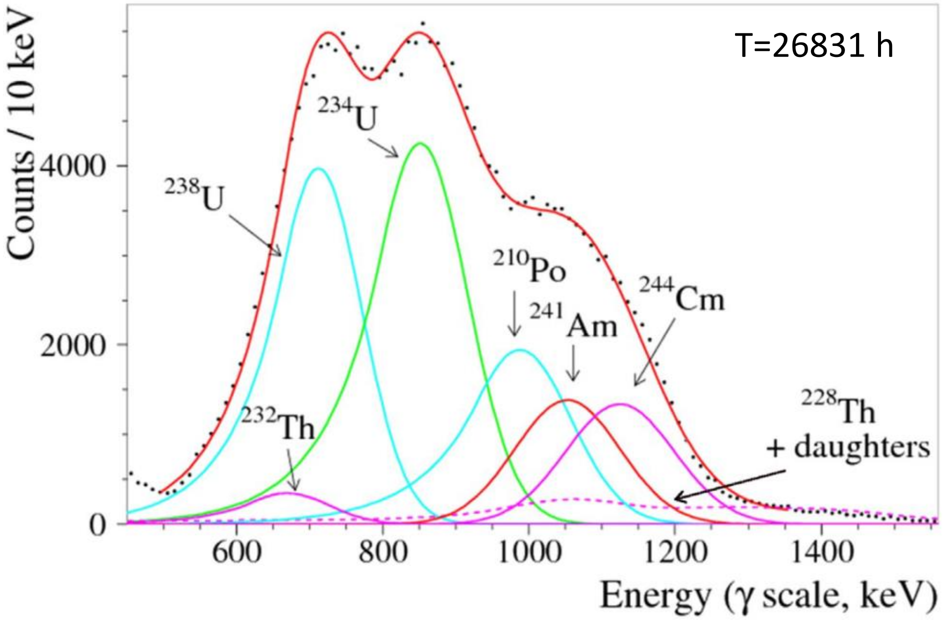
- α activity in crystals #1 = 1.8 mBq/kg
- α activity in crystals #2 = 2.7 mBq/kg

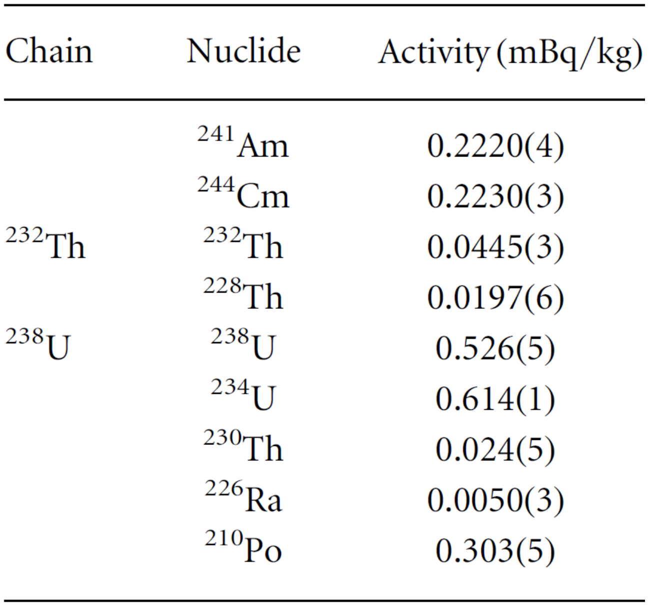
[1] AS Barabash *et al* 2016 *Nucl. Instrum. Meth. A* **833** 77

Data analysis: model for α spectrum

[P Belli *et al* 2022 *Phys. Scr.* **97** 085302]

The total α spectrum of the two $^{116}CdWO_4$ detectors (dots) was fitted by a model (red line) that includes the internal contaminations of the two scintillators (nuclides from U/Th families, ^{241}Am and ^{244}Cm) and the residual $\gamma(\beta)$ background





Chain	Nuclide	Activity (mBq/kg)
	^{241}Am	0.2220(4)
	^{244}Cm	0.2230(3)
^{232}Th	^{232}Th	0.0445(3)
	^{228}Th	0.0197(6)
^{238}U	^{238}U	0.526(5)
	^{234}U	0.614(1)
	^{230}Th	0.024(5)
	^{226}Ra	0.0050(3)
	^{210}Po	0.303(5)

Data analysis: the α/γ light ratio

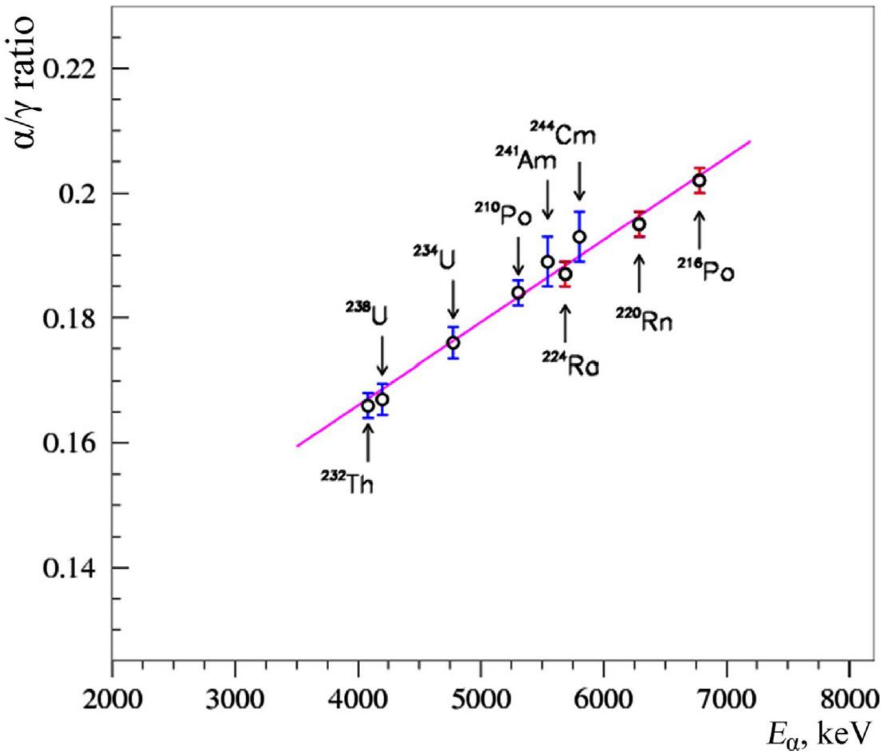
[P Belli *et al* 2022 *Phys. Scr.* **97** 085302]

α/γ light ratio is defined as the ratio of α peak position in the energy scale measured with γ sources to the energy of α particles

It has been estimated from the positions of the α peaks of the α active nuclides from U/Th chains, ^{241}Am and ^{244}Cm

In the energy interval [4.0, 6.8] MeV, the α/γ light ratio is:

$$\alpha/\gamma = 0.111(3) + 0.0133(5) \cdot E_{\alpha}[MeV]$$



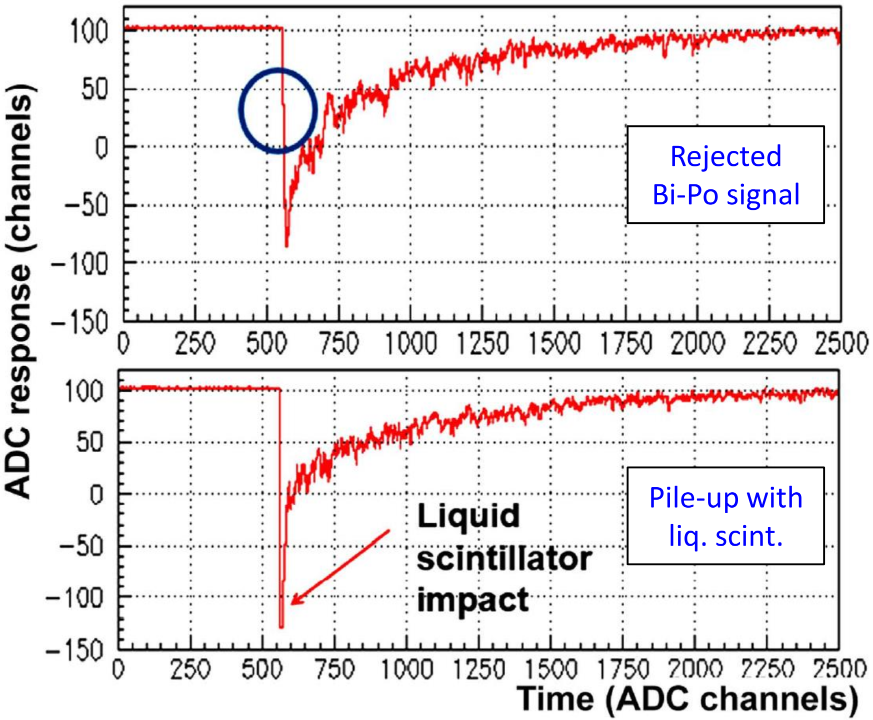
Data analysis: candidate events selection

[P Belli *et al* 2022 *Phys. Scr.* **97** 085302]

Selection of the candidate events from α decay of a daughter nucleus of long-lived Sg:

1. PSD+front-edge analysis used to reject:
   a) $\gamma(\beta)$ events
   b) Bi-Po events
   c) events with pile-up in liquid scintillator
2. Only high energy events, $E_\alpha > 8.9$ MeV, are considered (E_α evaluated with the α/γ light ratio)

$\Rightarrow$ 551 selected candidate events with E_α in [8.9, 14] MeV



Rejected Bi-Po signal

Pile-up with liq. scint.

Data analysis: result

[P Belli *et al* 2022 *Phys. Scr.* **97** 085302]

Fit of the α spectrum with a two-component model:

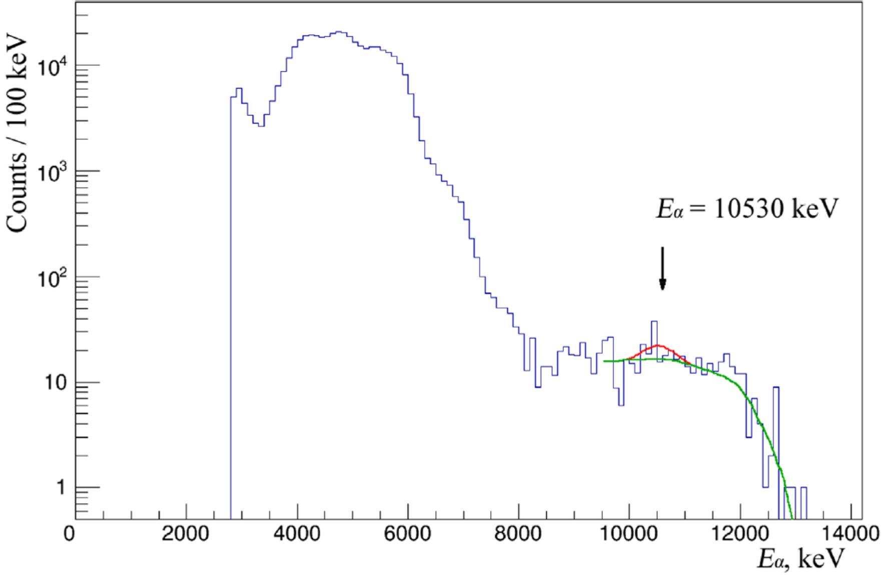
1. Simulated ^{212}Bi-^{212}Po spectrum (main background)
2. Searched α peak with variable position in [8.9, 14] MeV

⇒ Upper limit *limS* = 24.1 (90% C.L.) on the number of α decays of Sg's daughter nuclei, obtained as the maximal obtained value of the peak area

Then, considering:

- exposure collected with the two crystals: 36050 kg×h
- W atoms in the two crystals: $N_W = 2\times9.7\times10^{23}$
- efficiency for pulse-shape selection of α events ε = 96%
- assuming α detection efficiency ≈ 1
- assuming half-life for seaborgium $T_{1/2}(Sg) = 10^9$ yr

Energy spectrum of the α particles registered with the help of the two ^{116}CdWO$_4$ crystal scintillators (in α energy scale)



$$lim\left(\frac{N_{Sg}}{N_W}\right) = \frac{\lim S\cdot T_{1/2}(Sg)}{\ln 2\cdot\varepsilon\cdot N_W\cdot t} = 5.1\times10^{-15}\ \text{atom(Sg)/atom(W)}\ \ (90\%\ \text{C.L.})$$

Conclusions

- Assuming $T_{1/2} = 10^9$ yr for the Sg long-lived nuclide (standard assumption in the search for naturally occurring SHEs), an upper limit on atomic abundance of naturally occurring superheavy element Sg has been set to:

  5.1×10^{-15} atom(Sg)/atom(W) at 90% C.L.

- The limit is ≈11 times better than that obtained in a previous search with a $ZnWO_4$ scintillation detector [P Belli *et al* 2015 *Phys. Scr.* **90** 085301]

- The reached sensitivity is better than or comparable to that of other kinds of experiments which look for spontaneous fission of natural SHEs or use the accelerator mass-spectrometry