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ICP-SFMS SEARCH FOR LONG-LIVED NATURALLY-OCCURRING HEAVY, SUPERHEAVY AND SUPERACTINIDE NUCLEI COMPARED TO AMS EXPERIMENTS

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Negative results obtained in AMS searches by Dellinger *et al.* on mostly unrefined ores have led them to conclude that the very heavy long-lived species found in chemically processed samples with ICP-SFMS by Marinov *et al.* are artifacts. We argue that it may not be surprising that results obtained from small random samplings of inhomogeneous natural minerals would contrast with concentrations found in homogeneous materials extracted from large quantities of ore. We also point out that it is possible that the groups of counts at masses 296 and 294 seen by Dellinger *et al.* could be, within experimental uncertainties, due to ^{296}Rg and ^{294}eka-Bi in long-lived isomeric states. In such case, the experiments of Dellinger *et al.* lend support to the experiments of Marinov *et al.*

Keywords: Atomic mass measurements; long-lived isomeric states; superheavy elements; natural materials.

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In a recent paper entitled "Ultrasensitive search for long-lived superheavy nuclides in the mass range $A = 288$ to $A = 300$ in natural Pt, Pb, and Bi," Dellinger *et al.*[1] searched for superheavy elements, mainly in native samples of Pt, Pb, and Bi, using the AMS technique. Negative results were claimed in this and in their previous investigations on Th (Ref. 2) and Au (Ref. 3), with upper limits of relative abundances in the range of 5×10^{-13} to 10^{-16} g/g. The authors then concluded that the observation of long-lived isomeric states, by measuring accurate masses using ICP-SFMS, in various heavy, superheavy and superactinide nuclei are artifacts. Such isomeric states have been reported in the neutron-deficient ^{211,213,217,218}Th

isotopes (abundance $(1-10) \times 10^{-11}$ relative to ^{232}Th),[4] in the superheavy nuclei ^{261,265}Rg (abundance of $(1–10)\times 10^{-10}$ relative to Au),[5,6,a] and in the superactinide nucleus ^{292}eka-Th (abundance of $(1-10) \times 10^{-12}$ relative to ^{232}Th).[7]

An important difference between our works[4–7] and those of Dellinger *et al.*[1–3] is that we used processed Au and Th starting materials, and Dellinger *et al.*, except for a few cases, used raw minerals. It may not be surprising that results obtained with random samplings of a few mg would contrast with concentrations found in processed homogeneous starting materials which probe a large amount of natural ore. This factor could be more important than presumably very little differences in separation factors between an eka-element and its lower homologue that might occur during the purification of the lower homologue from the ore. The cases where Dellinger *et al.* studied processed samples were ThO_2,[2] Pt, PbS (galena) and Bi.[1] The latter three are irrelevant for comparison with our results since we did not measure these elements. (However see below.) As for ThO_2, we do not think, that based on a single measurement using a very complicated system, one can conclude that all our measurements[4–7] done with the relatively straightforward ICP-SFMS system are artifacts. It would be more convincing to point out a weakness in our measurements, which neither we nor Dellinger *et al.*[1–3] have been able to find.

It is claimed[2] that they checked the efficiency of their AMS system by measuring the ratio of ^{228}Th/^{232}Th. For ^{232}Th in equilibrium with its daughters this ratio should be equal to the ratio of the corresponding half-lives which is 1.4×10^{-10}. However, when one measures the mass 228 with AMS (or with ICP-SFMS) one measures, together with ^{228}Th, also ^{228}Ra, which belongs to the same radioactive chain. Its half-life is 3.0 times longer than that of ^{228}Th. In addition, it is possible that the formation of negative ions of RaO_2 is higher, perhaps much higher, than for ThO_2. According to the "Negative-Ion Cookbook" of Middleton,[8] the production of negative ThO_2 ions is quite poor, and the maximum current measured by him was 50 nA. It is not clear how Dellinger *et al.* obtained an average current of about 320 nA of negative ThO_2 ions (Table 2 of Ref. 2).

Another comment we would like to make is related to the conclusion of Dellinger *et al.* that based on their results, there are no naturally-occurring SHEs. In addition to what was mentioned above[4–7] and also in Ref. 9, it seems to us that even their results could indicate the contrary. We refer in particular to Fig. 4(b) in Ref, 3 on ^{296}Rg and Fig. 11(b) in Ref. 1 on ^{294}eka-Bi. Both spectra are clean, without pile-up. In the first one there is a group of five events and in the second one there is a group of six events, both very close to the estimated positions of ^{296}Rg and ^{294}eka-Bi, respectively. These groups were ignored by the authors on the basis of their measured residual energies. In the first case the peak appears at a residual energy of about 10.5 MeV, where according to the authors, its center should appear at 12.0 MeV. In the second case of ^{294}eka-Bi this peak appears at 11.8 MeV, where

[a]In Ref. 6 an enrichment of Rg relative to Au of three to four orders of magnitude has been achieved.

its calculated position should be at 13.0 MeV. Such differences of 1.5 and 1.2 MeV out of predicted energy loss in the detector window of 12 and 10.5 MeV (about half of the initial energies of the ions of 24.0 and 23.5 MeV, respectively) could be due to experimental and theoretical uncertainties. Besides in window thickness and energy calibration of the AMS detector, there could be uncertainties in the energy loss and range when extrapolated to unstudied heavy species like Rg and eka-Bi. In addition, the appreciable scatter of these ions in the detector window, due to their large energy loss, decreases the residual energy of the ions in the detector. In conclusion, a residual energy of 10.5 MeV instead of 12 MeV in the case of ^{296}Rg, and 11.8 MeV instead of 13.0 MeV in the case of ^{294}eka-Bi, when the total energy loss in the window is about 12 MeV, could be within the uncertainties inherent in the experiments.

If ^{296}Rg and ^{294}eka-Bi have been observed in these experiments, then it is a very important result. With $Z = 111$ and $N = 185$ for ^{296}Rg, $Z = 115$ and $N = 179$ for ^{294}eka-Bi, they are in the center of the island of stability predicted for nuclei in their normal g.s. Since if found in natural materials, their half-lives should be $\geq 10^8$ y, or otherwise they would have decayed away. However, the predicted half-lives for ^{296}Rg and ^{294}eka-Bi in their normal g.s. are 4.5×10^6 and 1.0×10^4 s, respectively.[10] A consistent interpretation is that, like in ^{211,213,217,218}Th (Ref. 4) ^{261,265}Rg (Refs. 5 and 6) and ^{292}eka-Th (Ref. 7) long-lived isomeric states exist in ^{296}Rg and ^{294}eka-Bi.

In summary, we have pointed out that the article of Dellinger *et al.*[1] does not show that the observation of long-lived isomeric states in neutron-deficient Th isotopes,[4] in superheavy ^{261,265}Rg nuclei,[5,6] and in the superactinide nucleus ^{292}eka-Th,[7] are artifacts. It is also pointed out that long-lived ^{296}Rg and ^{294}eka-Bi may have been observed by them. If so, based on lifetimes, it is argued that these species would not be in their normal ground state, but rather in long-lived isomeric states as have been reported earlier.[4–7] These results may add credibility to the original discovery of long-lived isomeric states in naturally-occurring heavy, superheavy and superactinide nuclei.

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