Search for Superheavy Elements in Nature (Experimental Approach)

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Abstract Attempts to find superheavy elements in Nature started in the middle of sixties when predictions on the possible longevity of nuclei stabilized by the closed $Z = 114$-proton and $N = 184$-neutron shells have been made. During the ensuing years hundreds of geological samples, their processing products and probes of meteorites were studied. In all experiments only upper limits of the superheavy elements concentration in the studied samples have been determined.

1 Background

The term "superheavy nuclei" was introduced by Wheeler in 1955 [1]. After extrapolation of mass formulae and analysis of the data on half-lives of heavy nuclei the author concluded, that one can "expect the existence of nuclei twice as heavy as known nuclear species" (up to $Z = 147$, $N = 500$), and that "the limits to the stability of *superheavy nuclei* are set primarily by neutron escape and by spontaneous fission.

The conclusion, that stability of the heaviest nuclei will be determined by the spontaneous fission, has been earlier drawn by Seaborg [2]. Half-lives of even-even nuclide (at the beginning of 50th only 10 of them were known) relative to spontaneous fission could be described by the Bohr's model [3].

The possibility to exist in Nature of superheavy elements (SHE)—elements heavier than ^{238}U depends on two determinatives:

- it is necessary, that one of superheavy nuclides would have a lifetime long enough to cross "half" the Galaxy (of about 2×10^4 y) to be found in cosmic rays, or that, comparable with the age of the Earth (of about 4–5 $\times 10^9$ y), to be found in meteorites or terrestrial samples;

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- in the Universe should exist a mechanism leading to the formation of superheavy elements.

2 Prediction of Existence of SHE

According to the liquid drop model, there should be the linear dependency between the logarithm of a spontaneous fission half-life and the fissility parameter - Z^2/A. Figure 1 shows the dependency of a log $T_{1/2}(s)$ on the fissility parameter Z^2/A (the dada were taken from [2, 4]).

From the Fig. 1 one can see the giant progress in studying heavy nuclei during past 60 years. The extrapolation of the general tendency of exponential decreasing of spontaneous fission half-lives toward the region of instantaneous fission with $T_{1/2} < 10^{-20}$ s has given a value $(Z^2/A)_{\mathrm{crit}} \approx 47$ [2].

It had become obvious, that departures of log $T_{1/2}$ by several powers of 10 from the linear dependence on the fissility parameter are principal. These deviations have been explained [5] by the suggestion of existence in the transuranium region of a "subshell" corresponding to $N = 152$ and being of a fundamentally different nature than the major closed shells.

The experimental facts indicating the influence of closed shells of $Z = 50$, 82 and of $N = 50$, 82, and 126 ("magic" numbers): numbers of isotopes and isotones, isotopic abundances, energies of radioactive decay, neutron capture cross-sections, were listed by Mayer [6].

For the first time "realistic" predictions of "magic" numbers beyond ^{208}Pb have been done by Sobiczewski, Gareev and Kalinkin [7] after analyzing single particle nucleon levels in a Woods-Saxon potential well. The possible magic numbers were found to be 114 and $N = 184$.

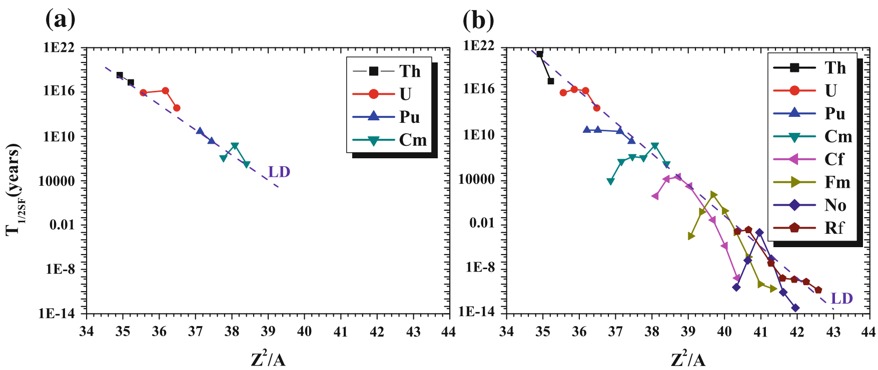

Fig. 1 Half-lives with respect to spontaneous fission as a function of the fissility parameter Z^2/A, **a** as it was in 1952, **b** recent data, 2012. The *dashed lines* represent a linear fit log $T_{1/2SF}$(y) $= 140.3 - 3.5 \cdot Z^2/A$

Theoretical predictions on the next closed shells numbers vary strongly depending on the model. Following the well-known proton and neutron shells with $Z = 82$ and $N = 126$ (^{208}Pb), the shell correction amplitude has a maximum for the superheavy nucleus $^{298}114$ at $N = 184$ in macro-microscopic models. After calculations performed using the Hartree–Fock–Bogoliubov (HFB)-model or self-consistent relativistic mean-field models, the closed spherical proton shells are predicted at $Z = 120$ or 126 [8].

Modern ideas of the influence of nuclear shells on stability of nuclei and results of calculations allow to conclude that the existence of superheavy nuclei with $Z \sim 114$ and $N \sim 184$ is reliably proved theoretically. Estimations of half-lives of such nuclei cover a wide range from fractions of a second till 10^{10} years and more.

3 Spontaneous Fission Properties of SHE

According to all theoretical predictions, superheavy nuclei self, or of their α- or β-decays will undergo spontaneous fission.

Probabilities of triple fission of nuclei with $Z \sim 114$ should be 10^3–10^4 times higher, than at fission of known nuclei [9]. According to [10, 11] the TKE of fission fragments of superheavy isotopes will make ∼300 MeV at fission into two, and ∼400 MeV at fission into three fragments.

Average number of prompt neutrons—$\overline{\nu}$ at fission of $^{298}114$ should be ∼11 [10]. Authors [11] estimated $\overline{\nu} = 8 - 10$. Kolb [9] concluded, that at a double fission $\overline{\nu} = 8$, however, at a triple fission $\overline{\nu} = 2 - 4$. Hoffman [12] analyzing correlations of mass distribution modes and average numbers of fission neutrons concluded, that at symmetric fission $\overline{\nu} \sim 6$, whereas at asymmetric one $\overline{\nu} \sim 12$.

Uncertainties in predictions should be taken into account at design of detectors for SHE searches.

4 Possible Mechanisms of Nucleosynthesis of SHE

In an explosion of supernova the prompt capture of multiple neutrons—r-process becomes possible.

Schramm and Fowler [13], Tonder [14] and Klapdor [15] have shown that at supernova explosions formation of nuclides with $Z \sim 110$ and $A \sim 300$ is possible. The relative abundance of superheavy elements in supernova products can reach $SHE/^{238}U = 0.1 \div 1$ [13].

To the opposite conclusion had been drawn by Howard, Nix [16] and Mathews, Viola [17]. According to [16, 17] nuclide with masses $A > 275$ cannot be formed in astrophysical r-process.

More recent calculations [18] revealed that the abundance of superheavy elements in the r-process can be comparable with that of uranium, but the yield of SHE depends strongly on forecasts of properties of $\beta-$delayed, neutron-induced, and spontaneous fission.

Modern ideas about mechanisms of nucleosynthesis do not forbid formation of superheavy elements. Due to explosions or "star-quakes" interstellar clouds could be enriched by heavy and superheavy elements. Probably such nuclei are a part of nuclear component of cosmic rays. Superheavy elements, could enter in processes of evolution of planetary bodies and be captured in some rocks or minerals.

5 Choice of Objects to Search for SHE

The choice of objects perspective for discovery of superheavy elements is based on results of predictions of their chemical and physical properties. Most predictions are based on extrapolations of properties within considered group of elements along with calculations of electronic configurations including relativistic effects [19–23].

Elements $113 \geq Z \geq 118$ should be, correspondingly, analogues of elements from thallium to radon, and a regular filling of the 7p-shell should occurs. The next noble gas, or to be more exact—a noble liquid with the boiling temperature of +15 C should be the element with $Z = 118$ [21]. Elements 119 and 120 should behave like alkaline metals. Starting with the element $Z = 121$ there should happen the filling of $5g$- and $6f$-electron shells, and elements $121 \geq Z \geq 154$ should form a series of superactinides [21].

Unfortunately, due to uncertainties of predicted position of the center of stability "island" ($110 \geq Z \geq 130$) one can consider as light chemical analogue of SHE almost any element from the Mendeleev's periodic system of the elements.

6 Experimental Search for SHE in Nature

The search for heavy elements in cosmic rays, terrestrial samples, and meteorites was in the 70's and 80's one of the extensive experimental investigations (see reviews [24–27]).

6.1 Search for SHE Nuclei in Cosmic Rays

Efforts of experimentalists were appreciably forced by Fowler's studies [28] of nuclear component of cosmic rays. In photoemulsion layers exposed at an upper atmosphere, two tracks have been found out, which were attributed to stops of nuclei with $Z = 90 \pm 4$. After recalibration of emulsions the authors [28] concluded, that the observed tracks corresponded to nuclei with $Z \approx 110$.

More perspective method of studying heavy component of cosmic rays has been proved by Flerov, Otgonsuren and Perelygin [29]. The method is based on revealing of traces from stops of nuclei in minerals from some meteorites, which could accumulate tracks during many millions years.

In olivine crystals from the Marjalahti meteorite [30] two tracks, attributed to nuclei with $Z > 97$, were observed. In a more recent study of Marjalahti meteorite [31] among 853 tracks with $Z > 50$, detected in 27 olivine crystals, 4 tracks, which could be attributed to the $Th - U$ group, were found. No tracks of heavier elements were observed.

The charge and energy of ultra-heavy nuclei will be measured using silicon detectors, aerogel and acrylic Cherenkov counters, and a scintillating optical fiber hodoscope in the frame of the ENTICE—the Energetic Trans-Iron Cosmic-ray Experiment) [32].

6.2 Study of Isotopic Anomalies

Accumulation of fission fragments of superheavy nuclei should lead to deviations in isotope abundances of known elements. In the seventies very popular was the hypothesis of Anders, Heyman [33], and Rao [34] about the origin of excess of heavy xenon isotopes in some meteorites—due to the spontaneous fission of superheavy elements. However, this hypothesis, despite its attraction, encountered a number of serious contradictions.

6.3 Study of Radiation Damages in Crystals

Decay of superheavy nuclei can result as well in radiation damages in crystalline media. It is known that at α-decay of isotopes contained in mineral inclusions, the so-called radioactive halos—spherical zones around a central mineral grain, will be formed. “Giant” halos having unusually large diameters corresponding to $\approx 14\,\text{MeV}$, an energy predicted for nuclides with $Z \sim 126$, were observed [35] in biotite from Madagascar. In order to identify elements around $Z = 126$ by characteristic X-rays, the inclusions—monazite crystals were irradiated by protons. In the first experiments [36] $L_{\alpha 1} X$-rays of elements 116, 124 and 126 were observed. Later, a more careful analysis and advanced technique for the excitation of X-rays [37] disproved the early results.

6.4 Mass-Separation of SHE

Several authors, e.g. [38, 39] tried to separate elements with masses $A = 250 - 350$ using mass-separation technique. Because of a background of molecular complexes of uranium and thorium the sensitivity of a “simple” separation method, as a rule, does not exceed 10^{-10}–10^{-11} g/g. Application of double mass-separation [40] allows to reduce considerably a background from molecular complexes, but reduces as well the overall sensitivity of this method.

Stephens [41] applied for studying of isotopic structure of platinum ore in experiments on searches for eka-Pt a more sophisticated technique—a tandem accelerator with a special beam line for ion analysis. This method is known as accelerator mass spectrometry (AMS).

In the latest experiments on the search for long-living roentgenium (Rg, $Z = 111$) isotopes in gold samples [42] using the VERA set-up, the sensitivity of 10^{-16} g/g was reported.

7 Direct Registration of SHE Decays

The most sensitive method of search for superheavy elements is connected with direct registration of their radioactive decays. At interpretation of results of such measurements one usually specify the limiting detectable dimensionless concentration C g/g of a searched element, assuming that their half-life makes $T_{1/2} = 10^9$ years. The searching methods based on detection of β-particles or γ-quanta can be ruled out due to extremely high background.

7.1 Study of Natural α-Activities

Experiments on studying weak natural α-activities were carried out for many years. A history of observations of 4.4-MeV α-activity over the period 1924–1979 in radiogenic haloes, zinc ores, monazite, thorite, huttonite, ultrabasic and other abyssal rocks, osmiridium, uranium ores, and raffinates of uranium is given in [43] (for more recent results see [42]). To search for superheavy elements in nature this method do not seems having prospects due to presence of numerous natural and man-caused α-emitters. The use of more advanced technique of detecting coinciding α-particles and characteristic X-rays [44] can give definitive results, but the overall sensitivity is insufficient for the searching experiments.

7.2 Study of Spontaneously Fissioning Activities

Essentially higher sensitivity of SHE searches provide methods based on detecting of spontaneous fission. This method appears as a universal one, because superheavy nuclides or the nearest products of their α-decay should undergo spontaneous fission. The whole accumulated experimental material allows to assert, that in nature exists only one spontaneously fissioning nuclide—uranium-238. Thus the problem of search of new elements by detecting their spontaneous fission can be reduced to reliable detecting of fission events and clearing up their possible origin due to uranium, contained in the sample.

A serious problem cause interactions of active cosmic radiation components—protons, neutrons and muons with heavy nuclei in probes or with detector parts. Whereas the nucleon component can be removed with a shielding of ≈10 mwe (meter of water equivalent, ≈4 m of concrete), for suppression of muons hundreds of meters shielding are needed. The fluxes of muons were measured in [45] up to 5200 mwe in connection with searches for rare processes.

7.2.1 Detecting of Fission Fragments

The first searches for SHE's spontaneous fission events were performed using solid state track detectors: Pb-glasses and glasses containing Bi, Tl, and W, which could accumulate tracks during $5 \div 200$ years [46]. As it was found out later [47, 48] with the use of sandwiched plastic track detectors, the additional background has been produced by cosmic rays.

Even higher sensitivity, but unfortunately also uncertainty, can be reached by etching internal fission tracks in suitable minerals, where they were accumulated over millions of years [49, 50].

Especially for the searches of rare spontaneous fission events of SHE, big proportional counters were developed [51]. The cathodes with an area of $\approx 1.6\,m^2$ were covered with a powdered samples layer, having the surface density $\approx 3\,mg \cdot cm^{-2}$. To reduce the "cosmic" background, the counters were operated in a room with a shield of 5 mwe.

A tricky detector - spinner [52] has been used for the observations of spontaneous fission of superheavy elements in chemical compounds, minerals, and in targets which were irradiated with 24-GeV protons [53]. The principle of operation of spinner is similar to that of bubble-chamber, but the negative pressure is produced in a working liquid by centrifugal forces. The main problem consist in the preparation of samples, which must be solvable in organic substances like ethanol.

7.3 Detecting of Prompt Fission Neutrons

The method, providing the highest sensitivity at the searches for SHE, is the detection of prompt neutrons accompanying fission. Practically all samples are transparent for neutrons, thus probes of up to 100 kg can be inspected. Another advantage is that there is no need to destroy the surveyed probe.

The advanced method of detecting spontaneous fission is based on detection of multiple neutron emission [54]. The detection of 2 and more neutrons unambiguously indicates the occurrence of spontaneous fission. Several kinds of neutron multiplicity detectors based on ^{3}He-filled proportional counters were designed at Flerov laboratory [55, 56].

This method provide for detecting spontaneous fission events the efficiency from 15–30 % ($\overline{\nu} \approx 2$) to about 50 % ($\overline{\nu} \approx 4$). The expected $\overline{\nu}$-values for superheavy elements were discussed earlier.

The major part of measurements was performed in the salt mine at a depth of 1100 mwe (passive shielding) with additional suppression of cosmic muons by active shielding based on Geiger-Müller counters. Long term measurements with high-purity samples (metallic lead, ferric oxide, melted quartz) showed that the "cosmic" background was less than 1 event per year. Thus, the only one source of background which should be considered is the spontaneous fission of uranium.

7.4 Search for SHE in Terrestrial Samples

At the first stage of searches several dozens of various ores and their processing products, products of metallurgy, ferro-manganese nodules, chemical preparations rich in platinum metals and gold, volatile metals such as Hg, Tl, Pb, Bi, rare earth elements were studied. The limits for the abundance of SHE were found to be less than 10^{-12} g/g [57, 58].

7.5 Search for SHE in Cheleken Peninsula Geothermal Waters

The heavy volatile metals content (e.g. thallium, lead, etc.) of the Cheleken Penninsula (the south-eastern coast of the Caspian Sea) water [59] is nearly 100 times that of oceanic water. These metals could escape from the Earth's crust, together with other volatile components. The extraction of heavy elements was carried out [59] using a vinyl-pyridine anion-exchange resin. Some 2,000 m^3 of the water were passed through a column containing 850 kg of the resin.

During a 88-day exposure of 9 kg of the saturated resin a total of 42 spontaneous fission events (0.5 events per day) have been recorded at neutron multiplicity detectors [60]. The analysis of the uranium content of the saturated resin, carried out using different methods ($(2-3) \times 10^{-8}$ g/g), had shown that the background due to spontaneous fission of uranium did not exceed 1 event from 42. Thus it was supposed that the detected spontaneously fissioning nuclide belongs to the region of superheavy elements.

Experiments were carried out aiming at the concentration of the detected activity by extraction of various chemical elements from 170 kg of the saturated resin. The fission counting rate of the obtained hydroxides was 5 counts per day, which corresponded to about 50 % of the initial spontaneous fission activity of the resin. It should be mentioned, that 1 atom of ^{252}Cf per 5 g of saturated resin could explain the observations. Further attempts [61] to concentrate the detected activity failed.

Table 1 Search for spontaneous fission in meteorites

Sample	^{238}U g/g	Weight kg	ε%	Time days	N of events n = 2	n = 3	n = 4
Saratov	$3.0 \cdot 10^{-9}$	5.2	22	94	4	1	0
Allende	$1.6 \cdot 10^{-8}$	3.9	22	40	3	0	0
Allende	$1.6 \cdot 10^{-8}$	22.5	12	55	10	1	0
Efremovka	$4.0 \cdot 10^{-8}$	11.7	12	105	14	1	0
Pb	$5.0 \cdot 10^{-9}$	100	22	15	0	0	0
Empty	–	–	22	200	0	0	0
Allende	$1.6 \cdot 10^{-8}$	10.5	30	45	5	2	1
SiO_2+MnO_2	$< 10^{-9}$	10.0	30	70	0	0	0
Pb	$5.0 \cdot 10^{-9}$	150	30	5	0	0	0
Empty	–	–	30	50	0	0	0

7.6 Search for SHE in Meteorites

Samples of meteorites "Saratov", "Efremovka" and "Allende" have been selected from the meteoric collection of Academy of Science of USSR. These meteorites belong to the class of chondrites and represent the less differentiated substance of the Solar system. The low uranium concentration in chondrites $3 \cdot 10^{-9}$–$4 \cdot 10^{-8}$ g/g allowed one to realize the maximum sensitivity.

Our investigations of meteorites were performed in 1972–1976 [62–64]. The results of measurements of spontaneous fission activity of meteorites along with background measurements with artificial samples analogous to these meteorites are presented in Table 1.

The observed counting rate was, on the average, 1 event per 5 days for 10 kg of a sample, what was $10 \div 30$ times higher than could originate from uranium contents and other background sources. Thus it was assumed that a long-lived spontaneously fissioning nuclide, possibly belonging to the region of SHE, is present in the studied samples of meteorites.

The isolation of the observed unknown nuclide (together with other volatile elements) has been performed by Zvara [65] from several kilograms of the Allende meteorite in hydrogen and then oxygen flows. Authors obtained an indication of its volatility in the elemental or oxidation state, but further attempts to increase the concentration of observed activity failed.

7.7 Discussion of Early Results

Figure 2 shows the correlation between the spontaneous fission activity observed in several samples, with the uranium concentration.

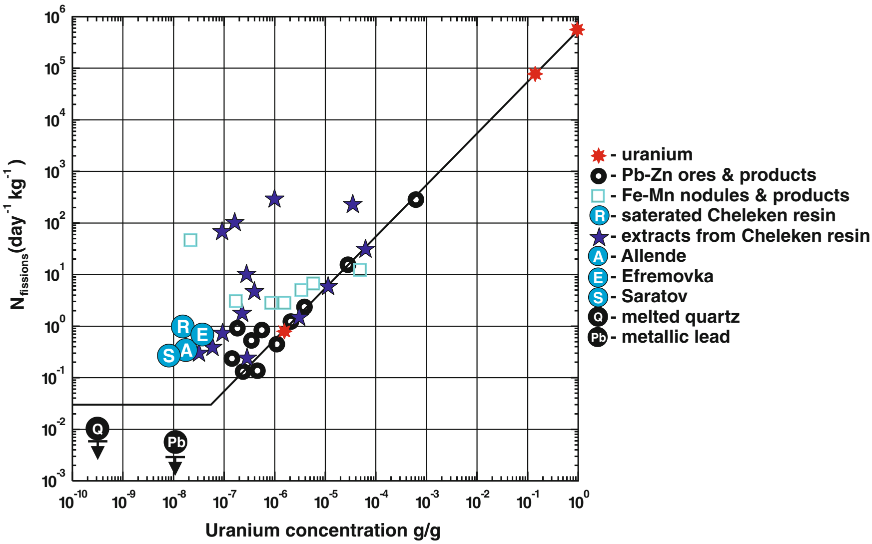

Fig. 2 Correlation between the uranium concentration and the spontaneous fission activity detected in the samples, recalculated to $events \cdot day^{-1} \cdot \mathrm{kg}^{-1}$

As one can see the counting rate for the majority of survived samples corresponds to the spontaneous fission activity of uranium. The activity in meteorites and products of processing of Cheleken geothermal waters exceeds that from uranium. But, also it did not show any clear correlation to known elements, and it did not follow them in attempts of further concentration.

In all experiments only upper limits of the superheavy element concentration in the studied samples have been determined. All this resulted in a pessimistic view on the possible existence of SHE in nature, and the searching experiments were practically stopped in the mid of 80-th.

8 New Samples for SHE Searches

The experimental data accumulated, and development of modern microscopic models during passed 30 years, simulated new approach to the search for perspective objects [66]. A noticeable increase in $T_{1/2}(\alpha)$ and $T_{1/2}(SF)$ may be expected for nuclei with $Z < 110$, which have not been yet looked for.

In accordance with the calculations [67], lifetimes of the isotopes $^{290-292}$Hs and $^{290-293}$Ds fall within the range $10^{10} \div 10^{14}$ s, and according to [68], these isotopes are β-stable (or have long lifetimes).

Considering different nuclei as objects for studies, it turns out that for element 108–Hs, the chemical homologue of Os, the chances to be found in terrestrial samples could be favorable. The search for rare decays may be undertaken with a metallic sample of raw Os.

Perspectives for SHE search, can follow from the discovered high volatility of Cn ($Z = 112$) and element $Z = 114$. These elements can be gases (noble) at normal conditions—the boiling temperature of Cn is (360 ± 100) K. Thus, e.g., one can look for SHE in heavy fractions of Xe production.

These experiments are running now with an especially designed neutron multiplicity detector [56] in the underground laboratory in Modane (France).

9 Conclusion

The problem of the existence of superheavy elements in nature belongs to the most fundamental, because it affects nuclear and atomic physics, quantum chemistry, astrophysics, cosmology, and undoubtedly the efforts to solve it will be continued.

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