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PERSPECTIVES OF THE SEARCH FOR SUPERHEAVY ELEMENTS IN NATURE

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The possibility of detecting long-lived superheavy elements in nature depends on the happy coincidence of two circumstances. First, there should be present an effective mechanism of nucleo-synthesis leading to the formation of the atomic nuclei of SHE with a fairly good probability. Secondly, it is necessary that these nuclei should include at least one beta-stable nuclide with a rather long lifetime for spontaneous fission and α-decay. If we speak about the search for SHE in the Solar System, this lifetime should be of the order of 10^9 years. But if we think about the existence of SHE in cosmic rays, half-lives of 10^6-10^7 years are admissible. The nuclear theory can predict such life-times for SHE only on the assumption that the parameters incorporated in it have extreme values. (Please refer to review paper [1] for the literature dealing with such predictions and other aspects of the SHE search problem).

As for the nucleosynthesis theory, it gives a number of restrictions on the probability of the formation of SHE nuclei. Moreover, there are some serious arguments supporting the assumption that this probability is vanishingly small in view of the cut-off of the r-process as a result of fission. Some authors are however inclined to think that this cut-off occurs at $A>300$. If this optimism proves to be justified, even then it is impossible to count on the fairly high abundance of SHE because of the average nonoptimal conditions of the r-process, large losses due to delayed fission in the decay back to the stability island and because of the radioactive decay of the longest-lived nuclide.

Searches for SHE in nature have a long-term history (see Fig.1), in the course of which their discovery was repeatedly announced erroneously. This circumstance, along with the unreliability of the theory, has borne a certain pessimism with respect to this problem. Despite this fact, we are actively engaged in this problem considering it to be very important and bearing in mind that no adequate experimental approach has been employed as yet to solve it.

Having at our disposal neutron multiplicity detectors surpassing similar devices in sensitivity by a factor of 10^2-10^3 we have detected in some primitive meteorites an unknown,

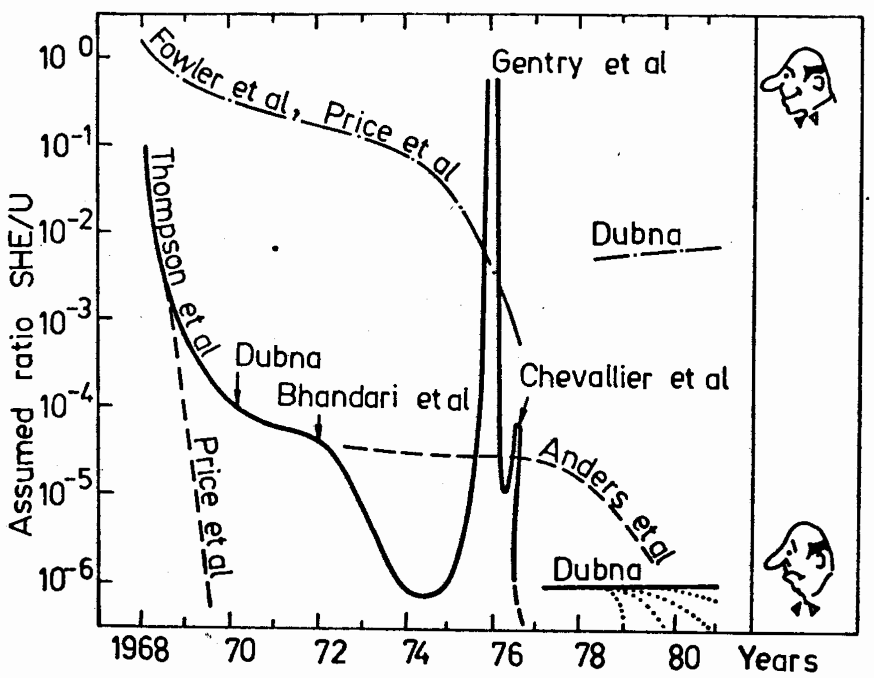

Fig.1. Retrospective view on the SHE searches. The concentration ratios SHE/U assumed by different authors are indicated. The dot-dashed lines correspond to cosmic rays. Full and broken lines stand for terrestrial samples and meteorites. Based on measurements of meteorites with neutron detectors we assumed in 1981 the ratio SHE/U $\simeq 10^{-6}$. The dotted lines reflect numerous attempts to extract SHE from Cheleken brines (see text).

spontaneously fissioning nuclide whose content amounts to 10^{-14} g/g [2]. If this result in fact indicates the existence of a long-lived superheavy element, the detection of this element in terrestrial samples, its extraction and concentration to the level required for its identification are very difficult tasks. This problem is simplified to some extent if hot spring waters are used as the initial object of research. The hot brines taken from Cheleken springs at a high general mineralization have a comparatively high content of heavy metals. By extracting from them the fraction of chemical elements heavier than iron we have detected a spontaneously

fissioning activity exceeding by several times the count rate for the activity of meteoritic samples [3].

Unfortunately the absolute value of this activity is very low: the count rate is approximately one spontaneous fission per day per ton of initial brine. Therefore the sought nuclide should be extracted from several tens and hundreds of tons of brine and concentrated in small samples by chemical processing. This extraction was made by different methods: cementation on metals, coprecipitation with hydroxides, sulfides and manganese oxide, ion-exchange concentration, extraction into the organic phase, and others. In the process of this work it was proved that the results of extraction of the activity are reproducible; however, nobody has so far succeeded in obtaining samples with a count rate that substantially exceeds one spontaneous fission event per day per kg. Evidently it is necessary to perform the multi-step concentration which needs the systematic control of numerous chemical fractions.

We believe that the conditions of such control can be satisfied by the mass-spectrometric method modified taking into account the modern technique of atom ionization in laser plasma and the technique of detecting ions, based on channel plates. We at Dubna are presently developing a mass-spectrometer based on three-step purification from the scattered ions of strong mass lines and including a lithium-vapor gas cell to eliminate charged molecules. By using this mass-spectrometer we hope to obtain the possibility of detecting 10^6-10^7 atoms of SHE in samples weighing about 100 mg. This will not only simplify the control of the SHE extraction process from hot brines, but also will make it possible to conduct a systematic search for terrestrial rocks and minerals that are rich in SHE relative to the primitive meteorites. In addition to the evident advantage of determining the mass of the nuclide sought, the mass spectrometer should facilitate the task of separating it from uranium and transuranium elements whose spontaneous fission background should be taken into account in the case of neutron multiplicity detectors very thoroughly.

Whether one can obtain reliable proof for the existence of SHE by establishing its mass and atomic numbers and other important physical and chemical properties will be clear in a not too distant future. However, the ambiguous results of extraction of the spontaneously fissioning nuclide from Cheleken brine cause our concern and impel us to conduct further searches for the background sources neglected. It is sufficient to say that the spontaneous fission activity of about one decay per day can be accounted for by the admixture of as little as 10 atoms of ^{252}Cf in each gram of the sample under study (10^{-20} g/g). Unfortunately, at present this possibility is not excluded completely.

The search for SHE nuclei in the cosmic rays looks promising because for comparatively young age (10^6–10^7 yrs) of the cosmic rays and the contribution of various sources of nucleosynthesis can result in a high content of the nuclei sought in the cosmic rays compared with the Solar System matter. This was first noticed by Fowler et al.[4], who has made the conclusion about the observation of one or two tracks of the $Z > 100$ atomic nuclei in emulsion exposed in the upper layers of the atmosphere. Subsequently, after a thorough analysis, the authors themselves demonstrated that those tracks had been caused by light element nuclei. A considerable amount of data on the charge spectrum of heavy nuclei has been accumulated by the two groups of Fowler[5] and Price[6] for more than 10 years of intensive studies using plastic track detectors and photoemulsion stacks exposed in the upper layers of the atmosphere in balloons and the orbiting station "Skylab". However, the statistics obtained in those experiments, in which a total of about 20 tracks of thorium-uranium nuclei have been observed is insufficient for SHE searches. In order to enhance the sensitivity substantially, it is necessary to expose in space detectors with an area of many dozens and even hundreds of m^2, and this is hardly practicable.

We at Dubna have decided to go another way by taking advantage of the capability of silicate meteoritic minerals (olivines) to record and conserve the tracks of atomic nuclei for several dozens and hundreds of millions of years. Mere estimates show that 1 cm^3 olivine located at a depth of less than 7 cm from the preatmospheric surface of meteorite must have accumulated 10^2–10^3 tracks of thorium-uranium nuclei for a period of 10^8 years. By using the appropriate etchant and providing the necessary conditions for the penetration of this etchant to the zones of latent defects it is possible to reveal in olivines visible tracks whose length is determined by the atomic number of the corresponding nuclei. The existence of the etching threshold for latent defects leads to the observation of but the final range of the nuclei (see Fig.2a). The etching threshold was increased artificially by the controlled annealing of crystals. Such annealing led to the shortening of the track lengths of the superheavy nuclei sought and to the removal of the smearing out of the track lengths caused by their fading for a long period of time. The shortening of the track lengths decreased the probability of their exit beyond the crystal volume. The annealing and calibration model experiments were performed by using the Cr , Fe ,

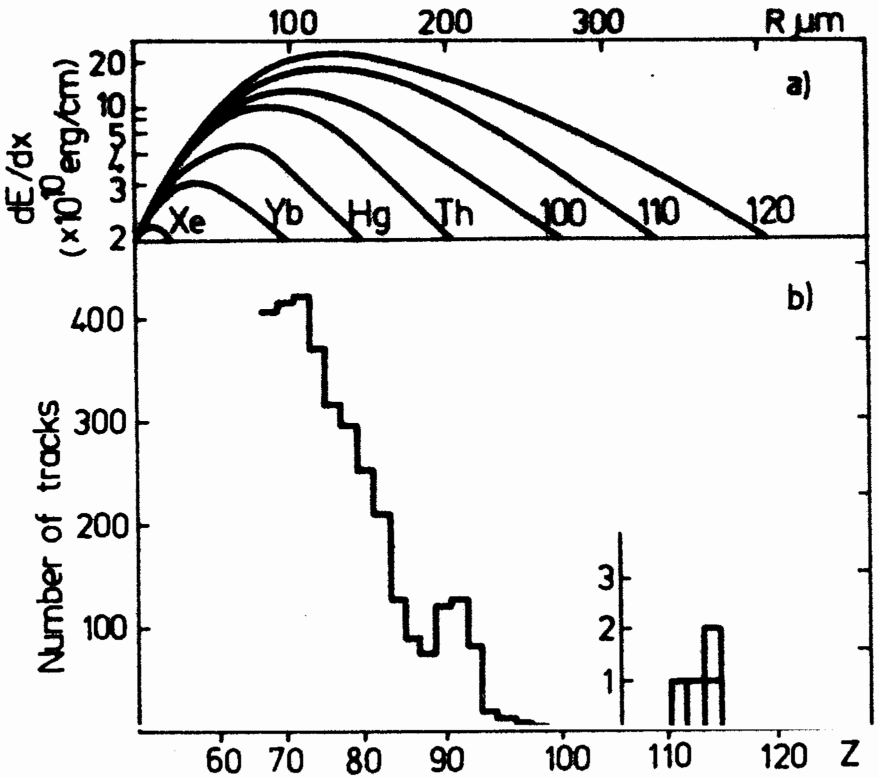

Fig.2. a) The dependence of the specific ionization of relativistic atomic nuclei on the residual range in olivine. $dE/dx = 2\cdot10^{10}$ erg/cm corresponds to the etching threshold for olivine after annealing for 72 hours at 380°. b) The track length spectrum for the $Z > 70$ nuclei detected in olivines from Marjalahti and Eagle Station meteorites. The abscissas of both plots (a and b) coincide. The atomic numbers are given at the bottom.

Ge , Kr, and Xe heavy ions. The experiments were made using olivines from the iron-stony meteorites Marjalahti, Eagle Station, Lipovsky Khutor and others.

Fig.2b shows the track length spectrum obtained as a result of scanning about 1.5 cm^3 olivine from Marjalahti and Eagle Station meteorites annealed for 32 hours at a temperature of 430°C[7]. This spectrum exhibits a group of tracks 180–230 μm in length which are obviously due to the thorium-uranium nuclei. Three tracks, 350 μm, >340 μm and 360 μm long, have been observed, which correspond to nuclei with $Z \approx 110$.

This assumption agrees with the results of comparison between the high-energy parts of these tracks and the thorium-uranium tracks. A part of the olivines was subjected to somewhat different annealing (at 380 °C, 72 hrs). The scanning of some of these crystals (0.3 cm^3) has led to the detection of about forty tracks of the thorium-uranium nuclei with a length of 380-440 μm that is expected for these nuclei under the chosen conditions of annealing. In addition, another, 710 μm track has been detected, which corresponds to a $Z \simeq 110$ nucleus. The estimates based on the data of Fig.2b and on other data not shown in this figure, give the ratio (SHE/Th-U $\simeq 3 \cdot 10^{-3}$) for the cosmic ray flux.

The volume of the olivines scanned can be increased by one order of magnitude. This will allow one to establish definitely whether or not SHE are present in the cosmic rays and to obtain reliable and detailed information about them. This hope rests on the following. First, owing to the existence of a wide interval between thorium-uranium and SHE one can hope that the SHE nuclei form a separate distinct group in the range of large track lengths. Secondly, in a not too distant future a possibility will arise to calibrate olivine crystals by lead and uranium ions, which will enhance the reliability of identification of new nuclei.

The statistics presented in Fig.2b exceed by approximately 70 times the amount of data obtained with the help of balloons and spacecrafts. Obviously the method developed at Dubna will have a crucial advantage in the nearest future in terms of the statistics collection rate. This method is characterized by another interesting feature. The radiation age of Marjalahti meteorite crystals is about 180 million years and they accumulate the tracks of the atomic nuclei of ancient cosmic rays. As 120 million years ago the Solar System was within the spiral arm of the Galaxy, where a higher frequency of Supernova explosions is observed, one can hope that olivines from Marjalahti meteorite contain the tracks of the atomic nuclei synthesized in those explosions. Taking into account the considerable range of the radiation ages of numerous meteorites, it is possible to carry out the differentiated search for the tracks of superheavy (and ordinary) nuclei depending on the place and time of their occurrence.

In the nearest future we are planning to extend the region of searches for SHE. This extension implies the search for SHE cosmic ray nuclei not only in olivines but also in other silicate minerals from meteorites. The SHE search in terrestrial samples will also be carried out using new geological subjects, the selection of which correspond to the various proposed mechanisms of geochemical migration of these elements.

Returning to the beginning of this paper I would like to note that the two essential problems on the solution of which the possibility of detecting SHE in nature depends are intensively studied by nuclear physics and astrophysics theoreticians. These studies can obtain a quantitative experimental basis if unambiguous and positive results are obtained in one of the directions discussed here. In our view, the cosmic ray studies are most likely to give such results. In this case we shall have an additional strong stimulus to the further searches for SHE in terrestrial samples as well.

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Флеров Г.Н. E6-82-32
Перспективы поиска сверхтяжелых элементов в природе

В связи с задачей поиска сверхтяжелых элементов обсуждается проблема концентрирования и идентификации спонтанно делящегося нуклида, обнаруженного в некоторых метеоритах и гидротермах. Представлены результаты поиска треков космических атомных ядер СТЭ в оливинах из метеоритов.

Работа выполнена в Лаборатории ядерных реакций ОИЯИ.

Сообщение Объединенного института ядерных исследований. Дубна 1982

Flerov G.N. E6-82-32
Perspectives of the Search for Superheavy Elements in Nature

The problem of concentrating and identifying the unknown, spontaneously fissioning nuclide detected in some meteorites and hot brines is discussed in the context of the search for superheavy elements. The results of searches for the tracks of the cosmic atomic nuclei of superheavy elements in meteoritic olivines are presented.

The investigation has been performed at the Laboratory of Nuclear Reactions, JINR.

Communication of the Joint Institute for Nuclear Research. Dubna 1982