

THE PHYSICAL AND CHEMICAL ASPECTS OF THE SEARCH FOR SUPERHEAVY ELEMENTS

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Abstract - The problem of the search for superheavy elements (SHE) in nature is considered on the basis of the studies performed at Dubna. The search for SHE nuclei in cosmic rays has been carried out. As a result of scanning 1.0 cm³ of olivine from meteorites, about 400 tracks of uranium-thorium nuclei have been observed. Two tracks have been revealed, which in length correspond to nuclei with $Z \sim 110$. The limit for the flow of magnetic monopoles in cosmic rays with magnetic charge more than five ($\leq 10^{-19} \text{cm}^{-2} \text{s}^{-1} \text{sr}^{-1}$) also follows from these data. Neutron multiplicity detectors have been developed, which allow one to detect a SHE admixture at a level of 10^{-15} g/g. In meteorites of the type of carbonaceous chondrites and in the water of the hot springs of the Cheleken Peninsula a spontaneously fissioning nuclide has been detected, which can apparently be assigned to the region of SHE. The irradiation of samples with ⁴He ions to observe the induced fission of SHE nuclei and the bombardment of these samples with resonant neutron beams to produce neighbouring short-lived nuclei have not given unambiguous results. Experiments are discussed, which should be performed for the identification of this nuclide. The possibility of performing an experiment on the search for nucleon decay with the violation of baryon number is also outlined. Neutron multiplicity detectors like those used in the search for SHE in nature can allow one to achieve the limit of a nucleon half-life of 10^{31} years.

The process of discovery of chemical elements covers many thousands of years and is one of the most important factors of the scientific progress which affects the level of productive forces and the spiritual life of mankind. The rate of the development of this process illustrated in Fig.1 has been governed not only by the quantity and quality of the accumulated just chemical information, but also, to a great extent, by the new research methods, which appear and are being developed in other fields of science (Ref.1).

In the 20th century nuclear physics has taken great interest in this "Glasperlenspiel" (see the novel "Das Glasperlenspiel" by Herman Hesse, 1943) using its new powerful methods based on the radioactivity registration, nuclear reactions, mass-spectrometry, etc. The peculiarity of this period is also the fact that by the beginning of the 50's, after the discovery of a number of elements (Hf, Re, At, Fr, Pa, Tc, and Pm) all the places in the Mendeleev Periodic Table turned out to be filled up to uranium, the heaviest of all known elements that has survived in the Solar System from the time of its formation. In this connection the question has naturally arisen as to whether any chemical elements heavier than uranium do exist in nature. No unambiguous answer to it has ever been obtained, although, as it seems to us, some results obtained at Dubna make it possible to reckon for its positive solution in the future.

The study of some problems of nuclear physics and other fields of physics and chemistry provides the still greater proofs for the fundamental importance of this question. We shall now try to make a short review of some of these problems. But before that we would like to refer to the permanent importance, which was always attached to the search and discovery of new elements. It manifested itself as the honour to assign the names to new elements presented to the authors of their discovery, who often commemorated in these names the names of great people, as well as the names of countries, regions or cities where they worked or were born. Such a principle has gained wide acceptance when designating the transuranium elements.

Beginning from the thirties, i.e., from the time of the discovery of a neutron, when it became clear that this particle permits the transformation of one atomic nucleus into another rather easily, the nuclear physicists became interested in the problem of artificial production of elements heavier than uranium. The attempts to perform such transformation of uranium are known to have resulted in the discovery of nuclear fission. Later the transuranium elements were synthesized, all the same, in reactions of neutron capture (Np, Am, Es, and Fm) and also in the course of bombardment of heavy elements with deuterons (Pu) and helium ions (Cm,

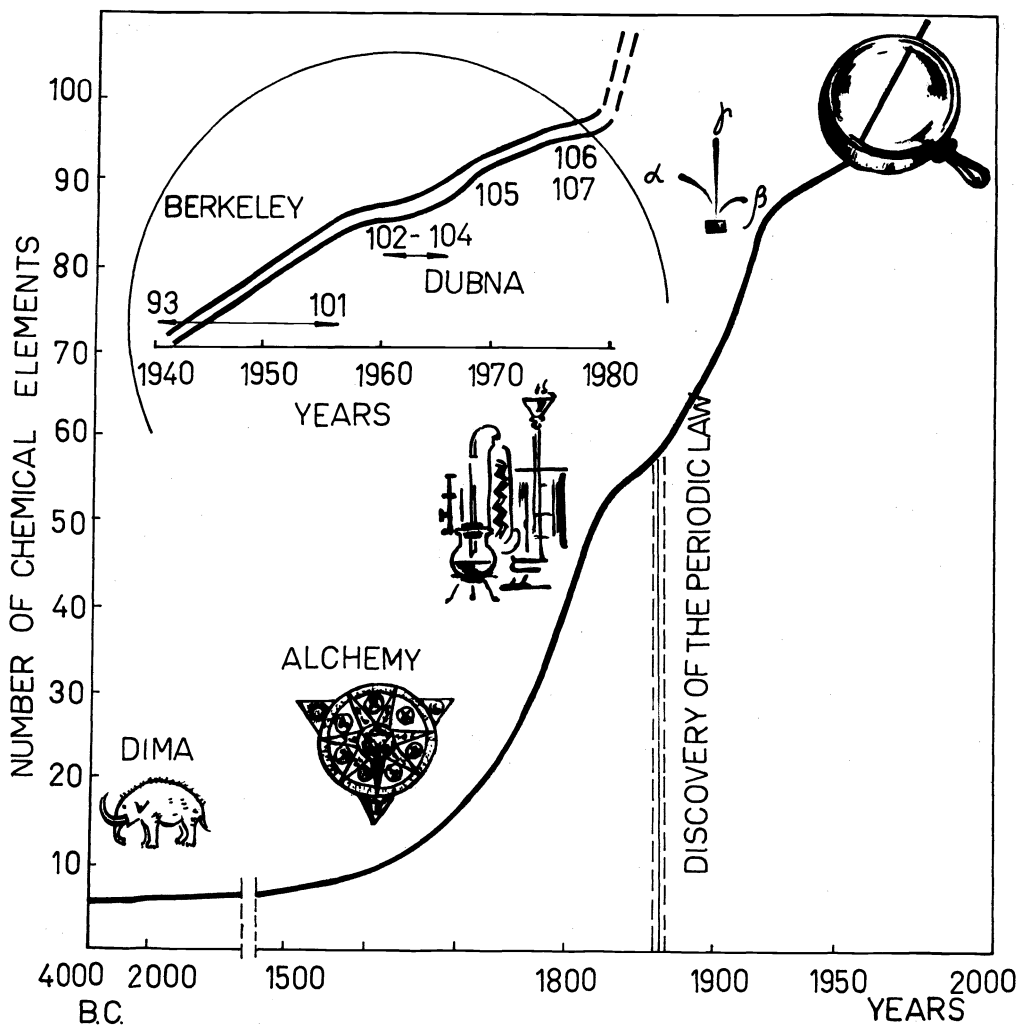


Fig.1. Number of known chemical elements vs time.

Bk, Cf, and Md). These extremely fine and now classical investigations (Ref.2) were performed by the group of American physicists and chemists in Berkeley headed by Seaborg, and they were of fundamental importance for the ascertaining of the fact of the existence of the actinide group and for the study of their chemical and physical properties. The study of the radioactive decay of these elements has left no hopes for their discovery in nature in any considerable amounts, since their lifetimes appeared to be too short compared to the age of the Solar System. It is true that ^{239}Pu isotope ($T = 2.4 \times 10^4$ years) produced in neutron capture reactions occurring under natural conditions, was found in uranium ores by Seaborg et al. (3,4) in experiments with a record, at those times, sensitivity. But its total amount in the Earth's crust is only several dozens of tons, which is much less than the amount of plutonium accumulated in nuclear reactions. In addition, in this case we do not deal with a chemical element which survived from the time of nucleosynthesis.

Subsequently it was necessary to make a choice between the two possible methods for synthesis of new elements - the one using the extremely intense fluxes of neutrons produced in thermonuclear explosions, and the other, by which the new nuclei are obtained in heavy ion reactions. The nuclear chemists of the USA, based on theoretical predictions and a number of other factors, gave preference to the first of them. Indeed, the einsteinium and fermium were first produced by this method. However, the program for the synthesis of new elements in underground thermonuclear explosions was brought to an impasse quite soon and did not result in any significant effects: the advance to the region with $Z > 100$ appeared to be impossible because of the instability of intermediate nuclides due to spontaneous and delayed fission.

The other method for synthesis, based on nuclear reactions with heavy ions was developed in the USA much less extensively. The accelerator HILAC constructed for this purpose (called SUPERHILAC after an improvement) was incapable of producing high enough beam intensity. In addition, the work at Berkeley was performed by practically one group. This resulted in a one-sided approach allowing neither to develop new specific methods, nor to study and eliminate the sources of the background. The underestimation in the USA of the method of detecting new

elements by spontaneous fission was particularly regretful. Therefore, the numerous essential mistakes could not be avoided in the results obtained by this group.

Experiments on the synthesis and study of the properties of the atomic nuclei of transfermium elements have been conducted at Dubna for more than 20 years. To perform these investigations, the JINR heavy ion cyclotron U-300 was put into operation in 1960. Using the advantages of this cyclotron, we were the first to obtain the correct data on isotopes of elements 102 and 103 and to discover elements 104 and 105 (Ref. 5). Somewhat later the work was completed in our laboratory, the results of which were the discovery of element 106 and certain indications of a short-lived isotope of element 107 (Ref.6).

In the studies of the properties of the newly synthesized nuclei the main attention was paid on the obtaining of the most complete information about their spontaneous fission, since this type of radioactive decay, discovered in 1940 (Ref.7) mainly sets the limits of existence of heavy chemical elements. The choice of this route and the development of the corresponding methods for identification of short-lived nuclei made it possible to obtain interesting results, both foreseen and unforeseen. Among the unexpected results are the discoveries of spontaneously fissioning isomers and delayed fission, which ultimately allowed one to obtain very important information on the regularities of nuclear fission, and could shed light on the problem of stability of heavy elements. What had been foreseen and was obtained was the systematics of spontaneous fission half-lives for heavy nuclides (Ref.8). These systematics reflected the marked, sometimes decisive effect of nuclear shells upon the smooth relationship between decay constants and the Z^2/A factor, which followed from the liquid drop model.

The tendency of half-lives to decrease with an increase in the atomic number of an element due to growing Z^2/A factor, dominates for all presently known transuranium elements. As a result, for nuclei with $Z > 100$ the role of spontaneous fission increases considerably compared to the α - and β -decay. While the ^{238}U isotope has a spontaneous fission half-life of about 10^{16} years (Ref.9), the most long-lived known isotopes with $Z = 104-106$ undergo spontaneous fission for about a second. The calculation of spontaneous fission half-lives is a very complicated task, which is virtually not yet solved up to now, in spite of the fact that many authors have been engaged in this problem. The outstanding paper by Wheeler (10) should be specially noted among the publications. In that paper, the conditions of the above problem were formulated and the possible existence of elements with atomic numbers up to 170 were first predicted on the basis of the liquid drop model. Using the more thoroughly developed liquid drop model and the semiempiric shell corrections to the nuclear binding energies Myers and Swiatecki (11) predicted the existence of considerable fission barriers for the atomic nuclei lying close to the magic numbers $Z = 114$ and $N = 184$, mentioned earlier by Meldner and Röpfer (12). Soon after that Strutinsky (13) proposed a method for calculating the shell correction to the binding energy of nuclei. This method played an important role in the understanding of the peculiarities of the fission barrier. The calculations first performed with the help of this method (Ref.14) indicated the possible existence of the whole series of atomic nuclei in the vicinity of magic numbers $Z = 114$ and $N = 184$, which formed a new island of relatively stable superheavy elements (SHE).

Later several authors performed the calculations of fission barriers and lifetimes for the unknown nuclei of SHE. The methods used for such calculations and their results were discussed in numerous original papers and reviews (see, e.g., Ref.15-18). Almost all authors come to the conclusion about the existence of one or more regions of superheavy atomic nuclei whose half-lives for spontaneous fission are many orders of magnitude greater than the characteristic nuclear time. The region of atomic numbers $Z = 110-120$ is mentioned in this connection more or less unambiguously. The half-lives of the most long-lived β -stable nuclides taking α -decay into account are estimated by various authors to lie between several hours and 10^{10} years. One can hardly hope for a decrease in this spread in the theoretical predictions in the nearest future, although an answer to the question as to whether or not SHE can be a subject of experimental investigations at all, depends crucially on how long they live.

As far as the possibility of the SHE discovery in nature is concerned, this seems to be very unlikely, since the happy coincidence of two circumstances is necessary for that. On the one hand, there should exist an effective mechanism of nucleosynthesis leading with sufficient probability to the formation of SHE atomic nuclei. On the other hand, there should exist at least one nuclide belonging to the new region of stability, which might have half-lives not shorter than hundreds of million years (in the case of the search for SHE in cosmic rays, a half-life of about 10^6-10^7 years is sufficient).

The probability for SHE to be generated in the course of the classical (or dynamical) r-process is apparently not high (Ref.19,20). The yield of these elements, as noted in the studies of Schramm's group (Ref.19) is, to a great extent, determined by the surface energy parameters of the nuclei, which are far from the β -stability line and through which the r-process passes. As a result of the errors in the surface energy parameters, one can predict with an approximately equal probability the yield of SHE to range from one tenth of the uranium yield to vanishingly small values. If the extrapolation leading to optimistic conclusions regarding SHE synthesis is valid, then one can always choose reasonable parameters for the r-process, resulting in the sufficiently high yields of nuclides of the new island of stability, which are of interest from an astrophysical point of view (i.e., those with half-lives $\geq 10^6$ years)

(Ref. 19). The numerical calculations of SHE yields in nucleosynthesis, presented in original papers give values between unity and 10^{-4} relative to the yield of uranium. Naturally, the lower limit of such estimates can be much lower than 10^{-4} due to the uncertainty of parameters.

Some authors (Ref.21,22) discussed the problem of the effect of delayed fission upon the yield of SHE in nucleosynthesis. It was noted that this phenomenon might lead to a considerable decrease in yields. However, such routes of sequential neutron capture alternating with β -transitions are evidently possible (Ref.22) when losses in delayed fission are not too high. For instance, this is possible for the so-called n-process (Ref.23).

It should be noted that the SHE nucleosynthesis mechanisms other than the r-process were also discussed in the literature. It is not excluded, for example, that a part of the Universe substance might have been synthesized in processes occurring in neutron stars (pulsars) (Ref.24).

Despite the low probability for SHE to exist in nature, 12 years ago we started to search for them and continue this work intensively, considering it to be of fundamental importance. The following circumstances inspire us to do that. First, it is not excluded that the search for SHE in nature is the only accessible way of detecting long-lived SHE and the only possibility to study thoroughly their physical and chemical properties. At any rate, only in this way we can reach the atomic nuclei lying in the centre of the island of stability. Secondly, even if the SHE are produced with heavy ion beams, it is clear already now that the most powerful accelerators (e.g., the U-400 cyclotron of the JINR) will be able of producing a maximum of only several new atoms a day. And if one succeeds in detecting at least one SHE isotope in the Earth's crust, then there will appear a possibility for its extraction in weighable amounts.

With targets containing 10^{12} atoms of a SHE, quite a number of new isotopes can be produced and studied, which belong to the island of stability (Ref.8). For instance, the modern facilities make it possible to perform continuous irradiations of such a target in the fluxes of $3 \times 10^{15} \text{ s}^{-1} \text{ cm}^{-2}$ thermal neutrons, $3 \times 10^{14} \text{ s}^{-1} \text{ cm}^{-2}$ resonant neutrons, $10^{15} \text{ s}^{-1} \text{ cm}^{-2}$ γ -rays, $10^{14} \text{ s}^{-1} \text{ cm}^{-2}$ T, ^3He , ^4He and other ions. New SHE isotopes, more or less distant from the initial long-lived nuclide, can be produced in sequential neutron capture reactions, in reactions (γ, n) , (γ, p) , (T, p) , $(^3\text{He}, p)$, $(^3\text{He}, n)$, and $(^4\text{He}, n)$, in the transfer reactions induced by heavy ions and in other reactions (see Fig.2).

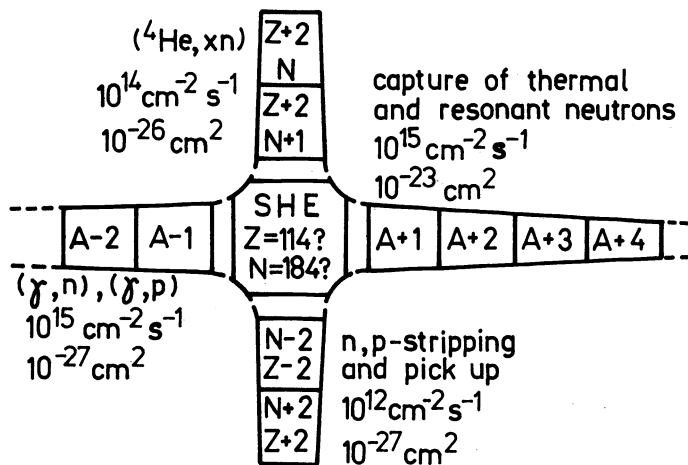


Fig.2. Illustration of the possible methods for synthesis of new atomic nuclei using targets of a naturally occurring SHE. Nuclear reactions, accessible particle and γ -ray fluxes and reaction cross sections are indicated.

Thirdly, the search for SHE in the Solar System and in cosmic rays can give important astrophysical information, in particular, on the physical nature of those cosmic objects in which, according to the modern concepts, nucleosynthesis is possible.

The history of the search for SHE is characterized by the periods of up's and down's in the activity of investigations. Initially, great interest in this problem was roused after the paper by Fowler (see Ref.25), in which he announced observation of one or two tracks of atomic nuclei with $Z \approx 110$ among the tracks of cosmic rays in the photoemulsions exposed at great altitudes. Later, as a result of the thorough analysis performed by the authors themselves, it was shown that, in fact, these tracks had been produced by the atomic nuclei of the lighter elements (Ref.26). The further rise of interest in the search for SHE in nature was caused by the communication (Ref.27) of an American group about the simultaneous observation of three superheavy elements in Madagascar monazites. As known, this result has not

been confirmed in the experiments performed using many methods in various laboratories.

The more than ten year period of search for SHE has shown that this task is an extremely complicated one and it requires great efforts to be made. Therefore, the majority of those who were initially attracted by the possibility of discovering a new island of stable elements, have abandoned this problem after one or two attempts made. And just only few groups are still engaged in this problem systematically and use increasingly sensitive and versatile techniques of the search. Nevertheless, more than one hundred experimental papers devoted to this subject have been published and in nearly the same number of papers some aspects of the experiments aimed at the search for SHE were discussed. There are excellent review papers by Herrmann (18,28), in which the detailed bibliography of relevant publications is presented. The theoretical and experimental aspects of the SHE problem as a whole, including the search for SHE in nature, were discussed at two Symposia (Ref.29,30).

Various authors proposed and developed different approaches to the search for SHE in nature, which should be considered with criticism from the point of view of their universality, sensitivity and feasibility within a reasonable period of time. Proceeding from such evaluation, we at Dubna give preference to the two directions - the search for SHE in galactic cosmic rays and the search for the spontaneously fissioning SHE (by detecting their spontaneous fission) in terrestrial samples and meteorites.

The existence of atomic nuclei with $Z > 30$ in cosmic rays was established in 1967 by the two groups: the group of Fleischer (88) and of Fowler (98). Since then, the study of the mass-spectra of these nuclei is continued by both groups (Ref.26,34). The recent experiments were performed at the cosmic station "Skylab" (Ref.35) and on a satellite (Ref.90). In these experiments the stacks of polymer films and photoemulsions were mainly used to detect heavy nuclei. For a decade of work in the cosmic space, a total of 30-35 m² of track detectors have been exposed (on the average, 3-3.5 m² per year). As a result, a total of 23 uranium-thorium nuclei have been detected and interesting information about the abundance of atomic nuclei in cosmic rays have been obtained. Although the authors of this work did not pursue the purpose of searching for SHE, they present some data on the limit of the flow of SHE nuclei in cosmic rays ($(Z \geq 110) / (74 \leq Z < 87) < 0.007$). However, the sensitivity of their experiments is undoubtedly not high enough for the search for SHE. To increase it considerably, it is necessary to expose in space for one year track detectors whose area is equal to hundreds or thousands of m². This is an extremely complicated problem since one should bear in mind the necessity of the thorough examination of the entire area of these detectors.

The studies of the mass spectra of heavy nuclei in galactic cosmic rays was carried out at Dubna with a view of searching for SHE. This work was carried out by Pereygin et al.(31,32) in collaboration with groups from Mongolia, France, Yugoslavia and India. The property (Ref.33) of silicate minerals, in particular, olivines, from meteorites to store and preserve the tracks of atomic nuclei during dozens and hundreds of million years was used in this study. Mere estimates show that in 1 cm³ of olivine hundreds or even thousands of tracks of the uranium-thorium group nuclei should be accumulated during this period of time. By using the appropriate etchant to reveal and enlarge the latent defects produced by heavy nuclei, one can obtain visible tracks, the length of which depends directly on the atomic number of the corresponding nuclei. This dependence can be obtained from the value of specific energy losses of the nuclei in olivine. The energy dependence of these energy losses, calculated for nuclei with different atomic numbers, is shown in Fig.3. The existence of the etching threshold for latent defects leads to the fact that only the end part of the range of relativistic nuclei, corresponding to maximal ionization losses, can be revealed. Atomic nuclei with $Z \leq 22$ do not leave any etchable tracks in olivine at all, since their ionization losses are lower than the threshold of etching at any energy.

For olivines studied at Dubna, calibration experiments with heavy ions from Ti to Xe were performed. The results of these calibrations and the obtained semiempirical Z dependence of the length of the etchable tracks of nuclei are summarized in Fig.3. As seen from this figure, tracks ~1400 μ m in length should correspond to nuclei with $Z \approx 110$, and the tracks due to uranium-thorium nuclei should be twice shorter. It should, however, be noted that the spectra of track lengths appear to be smeared, because during a period of 10^7 - 10^8 years the tracks are reduced due to the fading effect. The expected distributions of track lengths for some values of Z taking the fading effect into account are presented in Fig.4.

The smearing of peaks in the spectra of track lengths (see Fig.4) hinders the isolation of the maximum sought at $Z \approx 110$. Another difficulty is due to the relatively small dimensions of the available olivine crystals. The biggest of them can be obtained from the iron-stony meteorites (pallasites), with which all measurements have just been done. However, the average size of crystals even in these meteorites was 2-2.5 mm, which could result in considerable losses in track lengths exceeding 1 mm because they reach beyond the crystal boundaries.

By the partial thermal fading of latent tracks to cause the disappearance of their high-energy parts, one can improve the conditions of revealing the tracks of SHE nuclei. First of all, the spread in track lengths due to the difference in their age can thus be reduced greatly. The annealing model experiments were carried out for the tracks of heavy ions of Cr, Fe, Ge, Kr, and Xe. These experiments have shown that after the annealing the distributions of tracks have the form of narrow groups with a half-width of 3-5 μ m. Under the chosen conditions of

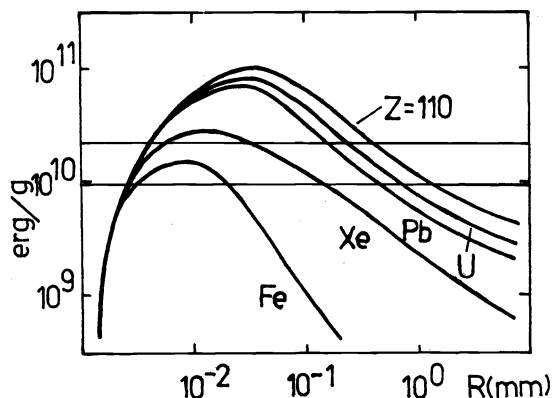


Fig.3. Dependence of the density of ionization losses in heavy ion energy in olivines on the residual range. The lower horizontal line corresponds to the threshold of etching for non-annealed olivines (10^{10} erg/g), the upper horizontal line is the threshold of etching (2.4×10^{10} erg/g) after annealing at 400°C during 32 hours. The etchable track lengths are equal to the lengths of the segments confined between the points of intersection of the curves with these lines.

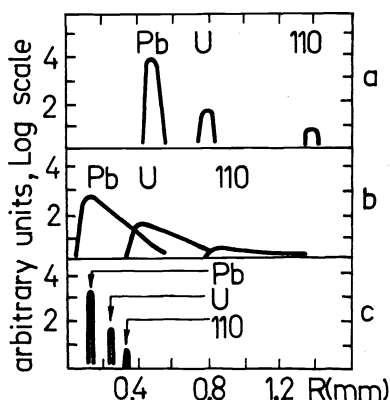


Fig.4. The distribution of track lengths for Pb, U and element $Z = 110$. (a) without fading, (b) with fading after 10^7 - 10^8 years in space, (c) after annealing at 400°C during 32 hours.

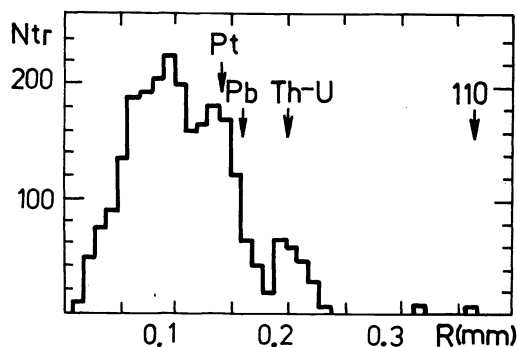


Fig.5. The spectrum of track lengths for very heavy nuclei ($Z > 70$) detected in olivines from meteorites.

annealing (400°C , $t = 32$ h) the etchable length of Xe tracks is $26 \mu\text{m}$. According to the extrapolation, such an annealing should result in the reduction of track lengths for uranium-thorium nuclei from $755 \mu\text{m}$ to 200 - $255 \mu\text{m}$ and from $1400 \mu\text{m}$ to 350 - $400 \mu\text{m}$ for the tracks of SHE nuclei (see Fig.3). Such annealing leads to the complete elimination of the background due to the tracks of the cosmic ray nuclei of the iron group ($Z \approx 26$), which facilitates substantially the search for the tracks of nuclei with $Z \geq 60$. Another advantage of annealing is a decrease in the track losses for nuclei with $Z \geq 90$, due to the exit of the tracks from the crystals.

During a short period of time, about 1 cm^3 of olivine crystals were examined and about 2500 tracks of nuclei with $Z \geq 70$ were measured. The spectrum of the track lengths for these nuclei, shown in Fig.5, exhibits a pronounced group of 180 - $230 \mu\text{m}$ tracks, which are evidently due to the uranium-thorium nuclei. The number of such tracks is about 400. Two tracks about 320 and $370 \mu\text{m}$ in length have been detected, which correspond to nuclei with $Z=110$. The comparison of the forms of the high-energy parts of these tracks with those of the uranium-thorium nuclei shows their similarity, which is an additional evidence in favour of such identification.

It should be noted that the annealing of crystals led to the elimination of latent tracks with specific losses below 2.4×10^{10} erg/g, which corresponds to hypothetical Dirac monopoles with magnetic charge $n < 5$. The assumption that these tracks belong to a magnetic monopole with charge $n \geq 6$ should also be rejected, since the specific ionization along their length is clearly not constant. Thus, the data cited allows one to set an upper limit of $10^{-19} \text{ cm}^{-2} \text{ sr}^{-1}$ on the flow of magnetic monopoles with magnetic charge $n \geq 6$ in galactic cosmic rays.

The preliminary estimates based on the data of Fig.5 give the ratio of flows ($N_{SHE}/N_{Th-U} \sim (1-3) \cdot 10^{-3}$). It is conceivable that the volume of the scanned olivines can be increased by one order of magnitude rather easily. This will enable us to establish positively whether the SHE nuclei are present in cosmic rays and to obtain the more reliable and detailed information about them.

We note that the amount of statistics presented in Fig.5, together with the data published before (Ref.31,32), is at least 30 times greater than that obtained with balloons and satellites (Ref.26,34,35,90).

Starting our investigations on the search for SHE in the Earth's crust and meteorites, we were fully aware that the solution of this problem demands the development of an extremely sensitive technique capable of detecting new elements at very low concentrations (10^{-12} - 10^{-14} g/g or less). In fact, the very first attempts to detect SHE have already shown that their abundance is several orders of magnitude lower than that of uranium, which is equal to 3×10^{-8} g/g for the Solar System. This is not surprising in the light of predictions of the nucleosynthesis theory. The possible decay of the most long-lived nuclide of the SHE region during the existence of the Solar System should also be taken into consideration.

The analysis of various methods (Ref.36,37) has shown that the highest sensitivity, versatility and flexibility of the methods based on detection of spontaneous fission make these methods most preferable at least at the first stage of the search for SHE. The high sensitivity of these methods is provided by the fact that only one spontaneously fissioning chemical element - uranium - exists in nature. Therefore, as a first approximation, the task of searches reduces to detection of the rare events of spontaneous fission and to the understanding of their correlation with uranium decay or other possible sources of the background. The versatility of this method results from the fact that if not the long-lived SHE per se, then their daughter products should decay by spontaneous fission. The maximum sensitivity is achieved for those isotopes, which have lifetimes of about 10^9 years, that is close to the age of the Solar System. Isotopes with much shorter lifetimes could not have been preserved in nature. The experimental results are usually interpreted in terms of the concentration of the SHE sought in grams per gram, assuming that its half-life (T) for spontaneous fission is equal to 10^9 years.

The first experiments aimed at the search for the rare events of spontaneous fission were performed at Dubna with the help of solid state track detectors. Much attention was paid to lead glasses (Ref.38) and glasses containing Bi, Tl, and W (Ref.39), which being quite old (5-200 years), could accumulate the tracks of the SHE fission fragments. The source of the background of fission fragments was, in this case, not only the spontaneous fission of uranium admixture in the glasses, but also the induced fission of Bi, Pb, Tl, and W by cosmic rays (Ref.40). In the majority of the samples investigated, no tracks of fission fragments have been detected, which could be related to the decay of SHE (the concentration limit, $C < 10^{-13}$ g/g). However, for some lead glasses an excess activity over the background has possibly been observed, which corresponds to a SHE concentration of 7×10^{-13} g/g. The extension of this work was associated with certain difficulties involved in the passive registration of fission fragments, such as the limited choice of samples, their small weight, and specific chemical composition, and the impossibility of the accurate estimation of the background.

Therefore, we began to develop the active methods of detecting the rare events of spontaneous fission, which could enable us to study the various natural samples. The large proportional counters for fission fragments and neutron detectors were taken as a basis (Ref.36). Proportional counters consisted of steel or aluminium tubes 2 m in length and 25 cm in diameter. Their inner surface (1.6 m^2 in area) was covered with a thin layer ($3-3.5 \text{ mgcm}^{-2}$) of the preliminarily ground samples under study. A Nichrome filament $50 \mu\text{m}$ in diameter served as an anode. The counters were filled with methane, and the factor of gas multiplication was equal to 300 at 1200 V applied to the filament. The simplest electronic circuit was fed from accumulators.

The sensitivity limit of this device in the search for SHE was determined by the level of background from spontaneous fission of uranium, by the background from cosmic ray induced fission of Th, Bi, and Pb and other heavy elements contained in the samples under study, as well as by the intrinsic background of the device. To eliminate the background from cosmic rays the counters were placed into a room shielded with concrete walls 2m thick. The intrinsic background of the counter was measured to be not higher than 1 pulse per year. As a result, the only factor that limits the sensitivity of measurement was spontaneous fission of uranium. With a uranium concentration in the sample under study of about 10^{-6} g/g the limit for detecting SHE could be achieved, which corresponded to a concentration of 10^{-13} g/g.

A still higher sensitivity to SHE was achieved with the help of the detectors which registered prompt spontaneous fission neutrons. We used several different neutron multiplicity detectors (Ref.36,41-45). We shall describe in brief the principle of their operation and give the basic parameters of the last modification. 56 proportional counters filled with ^3He at a pressure of 7 atm were placed in a moderator unit (paraffin or Plexiglass) around the sensitive volume with the samples under study. The neutrons emitted from the sample and slowed down were registered by the ^3He -counters with high probability. The maximum probability of detecting the neutrons emitted from the centre of the 10-liter sensitive volume was

$\epsilon = 50\%$. The average lifetime of neutrons in the detector was $\tau = 20 \mu\text{s}$. The characteristic feature of spontaneous fission was the observation of events with two, three or more neutrons detected during a time interval of $(4-5)\tau$. The start-up of the circuit responsible for the search for such events occurred each time when a single pulse appeared in any of the ^3He -counters.

To eliminate the cosmic ray background, all the measurements were performed in a salt mine at a depth of 1100 m.w.e. with an additional suppression of the background from cosmic muons with the help of a system of Geiger-Müller counters. The background of single pulses caused mainly by α -decay of the uranium and thorium that entered into the structural material of the ^3He counters, did not exceed $2 \times 10^{-2} \text{s}^{-1}$. However, already for double events the background was negligibly small. For several years, during which measurements with different model samples of high purity (melted quartz, ferrous-ferric oxide, Plexiglass, metallic lead) were performed regularly, not less than 2 days a week, the limiting level of this background was obtained to be less than one event per year.

With the described method of observation of spontaneous fission, the detection efficiency depended on the average number of prompt fission neutrons and ranged from 25% ($\bar{\nu} = 2$) to $\geq 50\%$ ($\bar{\nu} \geq 4$). Thus, with samples 20 kg in weight, having registered 10-20 events per year, one could observe SHE at a concentration of 10^{-15}g/g . It is clear that with such a counting rate it is necessary to consider thoroughly the background from spontaneous fission of the admixtures of uranium and transuranium elements, since this counting rate (10-20 counts per year) might be caused by the presence in the sample of 10^{-8}g/g of uranium admixtures or by a contamination with some isotopes of transuranium elements. Among such man-made isotopes, the most dangerous is ^{252}Cf which has a half-life of about 2.6 years and undergoes spontaneous fission with ~3% probability. The presence of 1 atom of this isotope in 100 g of the sample is enough for the appearance of the background, which will complicate the search for SHE. The detection of such a small admixture of ^{252}Cf is naturally a very difficult task.

Unfortunately, in recent years specimens of ^{252}Cf were used in many laboratories, which was not always necessary. Due to aggregate recoil, such specimens produce contaminations which although safe biologically, complicate enormously the study of spontaneously fissioning nuclei. For example, at Berkeley it was one of the factors which excluded the identification of new elements by detecting spontaneous fission. We at Dubna have taken all the measures to preserve "the californium sterility" of the laboratory. For this purpose we excluded the use of open sources of ^{252}Cf and other actinide isotopes, for which the α -decay-to-spontaneous fission ratio is $< 10^5$. The regular control of the purity of the laboratory rooms was performed with the help of solid state track detectors of fission fragments.

The concentration of uranium in the test samples was measured by irradiation with resonant neutrons and by the determination of the ^{239}U and ^{239}Np yields. The additional control was performed using the methods of uranium determination by thermal-neutron induced fission and by the α -activity of the ^{210}Po sublimated from the samples. The possible contamination with Cm-Cf isotopes, used in our laboratory as a material for cyclotron targets, were checked by measuring the α -activity of the test samples. Both the chemical separation of the above elements and the α -spectrum measurements for samples 0.1-0.2 g in weight with an ionization chamber with a cathode 0.4 m^2 in area were conducted. Under these conditions it was possible to detect an admixture of Cm-Cf isotopes, for which the ratio α/SF was larger than 200. Since this ratio was not lower than 2×10^5 for the target materials used, such contamination could be easily controlled. For a mixture of transuranium elements from nuclear tests, which could be present in the samples under study, the α/SF ratio is 4×10^7 (Ref.46), which enabled us to exclude this source of contaminations completely.

It is of some interest to compare the sensitivity of our technique developed for the search for SHE with the sensitivity of some of the best ones developed by other authors. The Walker group (47) used the stacks of polymer films with a total area of up to 10 m^2 to detect spontaneous fission fragments (they also considered the possibility of increasing the area to 100 m^2). The authors of this publication reported that they could achieve the sensitivity for the SHE detection at a level of 10^{-15}g/g and pointed out that it was possible to measure with their technique the nuclear charges of fission fragments within 2 units, as well as to detect ternary fission. In our opinion, the use of the above technique is somewhat complicated for the SHE search in a large number of samples. On the other hand, it can be useful in the studies of fission of the already discovered SHE.

Certain advantages for the search for SHE are offered by the study of tracks accumulated in natural minerals (Ref.48-50). Price, Fleischer and Woods (49) and Haak (55), having studied some specific minerals presented the limiting data on the concentration of eka-lead to be $3 \times 10^{-15} \text{g/g}$, and eka-bismuth - $3 \times 10^{-17} \text{g/g}$. It seems to us that these results should be interpreted with certain caution since one should bear in mind the possibility of natural decontamination (also from SHE) of the naturally occurring minerals.

Some authors (Ref.51-53) considered the possibility of SHE discovery in natural samples by analyzing the xenon isotopic anomalies, i.e., the excess of ^{136}Xe resulting from the symmetric fission of SHE. In particular, the Anders group (51,52) proposed an explanation of the origin of such anomalies in the meteorites of the carbonaceous chondrite type as being due to the decay of SHE. We think that this method (especially for "extinct" chemical elements)

is an indirect one and the results of its application are difficult to interpret (ref.54-57). The interpretation of the results of Anders et al. (51-52) as being related to the discovery of SHE can in our opinion be considered only as a speculation.

The research groups at Berkeley and Oak Ridge used neutron multiplicity detectors to search for SHE (Ref.58-63). The disadvantages of their setups, i.e., the high γ -ray background (Ref.58), the insufficient shielding from cosmic rays and, apparently, the unsuccessful selection of structural materials (Ref.59) results in the fact that the sensitivity achieved was at least one hundred times lower than at Dubna.

A neutron multiplicity detector was also used by the group of Brandt in Marburg (64,65,81). This group did not use any massive shielding from cosmic rays. The samples under study were placed in contact with a surface-barrier detector of fission fragments. Pulses from this detector triggered the circuit which counted the prompt neutrons detected. Fission fragments were also recorded with a solid track detector; for this purpose, mica was used as a support for the samples. It is natural that the samples not exceeding several dozens of milligrams in weight could be studied under such conditions. Therefore, the separation of various fractions of SHE from massive natural samples was carried out to achieve high sensitivity. This seems to be a weak point of this method since it should be acknowledged that it is practically impossible to avoid considerable losses during the chemical isolation of SHE from complex natural mixtures.

Among the methods of the search for SHE, which do not imply detection of spontaneous fission is the method of mass-separation followed by the observation of the induced fission of SHE. This method was developed by the Orsay-Warsaw group (Ref.66) and used also in Julich (Ref.67). The sensitivity limit achieved was 10^{-11} - 10^{-13} g/g. Epherre and Klapisch (68) proposed to use two-fold mass separation, which permitted a considerable improvement of this limit. However, this method has so far been used in only one case (Ref.69) to study a sample of Madagascar monazites.

The comparison of the neutron multiplicity detectors and proportional counters operated at Dubna, with the devices and methods used in other laboratories suggests the considerable advantages of our setups, which enable us to conduct the systematic search for SHE in a large number of natural samples with an average sensitivity exceeding by one to three orders of magnitude the limit achieved by other groups.

Since almost all available terrestrial samples had undergone a complicated geochemical evolution, the selection of prospective samples was not simple. At the first stage of the work, we studied more than a hundred of various ore samples and the products of their processing, including both the by-products of metallurgy and comparatively pure chemical preparations and metals. In the samples of such type, the limits for the contents of SHE - the chemical analogs of the elements of almost all groups of the Periodic Table - were set to lie between 10^{-12} and 10^{-13} g/g. The Pb-Zn ores (Ref.70), ferro-manganese nodules (Ref.71) and hot spring waters (Ref.72) were studied most thoroughly. Subsequently we thought it appropriate to pay attention to meteorites belonging to carbonaceous and unequilibrated chondrites, which are known (Ref.73,74) to be the least differentiated formations of the Solar System and are not depleted of heavy volatile metals (Hg, Tl, Pb, Bi) - the homologs of which can presumably be the expected SHE.

Our investigations of meteorites were begun in 1972 and continued until 1976 (Ref.43,44,75,76). These investigations have received a great support of the Directorate and Dr.R.Clark of the Museum of Natural History of the Smithsonian Institution (Washington, USA), who provided us with the Allende meteorite samples.

As a result of thorough measurements, we have observed in the samples of the above meteorites the effect of multiple neutron emission, which was absent in artificial samples analogous to these meteorites in their basic composition. The observed counting rate was, on the average, one event for five days for samples 10 kg in weight, and this was 10-30 times the counting rate that could be explained by the uranium contents (2×10^{-8} g/g) of these meteorites and by other sources of background. To interpret this result we assumed that a previously unknown long-lived and spontaneously fissioning nuclide was present in the studied samples of meteorites. Under the assumption that this nuclide belonged to SHE, Zvara et al. (76) performed its isolation (together with other volatile elements) from several kilograms of the Allende meteorite in hydrogen and then oxygen flows. They obtained an indication of its volatility in the elemental or oxidation state.

For a more reliable identification and study of the properties of the new emitter, including the assignment of its atomic and mass numbers, it was necessary to produce it in sufficiently large quantities and to concentrate it in samples of small weight. Therefore, we started new experiments with samples of terrestrial origin. As the basic sample used in this study we chose the water of hot springs of the Cheleken Peninsula (South-East of the Caspian Sea), which are rich in heavy volatile metals (Tl, Pb) that are released together with volatile components from the comparatively little differentiated substance of the Earth's mantle (Ref.77). These elements are captured by sodium-calcium chloride brines in the zones of the deep ruptures of the Earth's crust, which are traced in the Cheleken Peninsula. The contents of heavy volatile metals (Ref.78) in the water of Cheleken boreholes is approximately 100 times higher than in oceanic water, the total mineralization being 200 g/kg, which is 10 times

larger than that of oceanic water. In the initial experiments, the concentration of elements heavier than iron was performed by using the vinyl-pyridine anion-exchange resin. About 2000 m³ of the water was passed through a column containing 850 kg of the resin. In the course of the 88 day exposure, 42 events of multiple neutron emission, mainly double and triple, were registered by the neutron multiplicity detector for a sample of saturated resin 9 kg in weight (Ref.45).

The multiplicity distribution of the events detected has made it possible, in principle, to estimate the average number of prompt neutrons $\bar{\nu}$, assuming that the ν value has a normal distribution with the σ_{ν}^2 variance. With the statistics available, the accuracy of such estimates was unfortunately not high. Thus, for meteorites the admissible range of $\bar{\nu}$ estimates was 2-6 (Ref.79). For the samples obtained from Cheleken brines, the best estimate yielded $\bar{\nu} < 3$ and $\sigma_{\nu}^2 > 2$, but the measured distribution in multiplicity was still very similar to that expected for ^{252}Cf ($\bar{\nu} = 3.756$, $\sigma_{\nu}^2 = 1.57$). Therefore it was necessary to conduct the direct determinations of the possible Cf admixtures in the samples under study (Ref.45,80). These determinations performed by measuring the activity of the Cm-Cf chemical fractions, showed that the admixtures of the isotopes of these elements, which could in principle get into the Cheleken water with atmospheric falls (nuclear tests) or as contaminations in the course of laboratory processing, were at least 100 times lower than required to explain the observed counting rate. It was, however, impossible to exclude completely the explanation of the observed activity as being due to a very small admixture of the pure ^{252}Cf isotope, for which the ratio of the probabilities of α -decay and spontaneous fission ($\alpha/\text{SF}=30$) was lower than the level of detection sensitivity.

Nevertheless, we have advanced the hypothesis that both in the products obtained from the hot spring waters of the Cheleken Peninsula and in meteorites we have observed the spontaneous fission of a previously unknown nuclide belonging to the SHE region.

The further experiments presented a non-trivial task, since the concentration of the spontaneously fissioning nuclide in the studied samples was very low - $\sim 10^{-14}\text{g/g}$ in meteorites and $\sim 3 \times 10^{-14}\text{g/g}$ in saturated resin. Therefore, it was first necessary to increase this concentration. In experiments with saturated resin (Note a) the successive precipitation with hydroxides and sulfides was used. Samples up to 10 g in weight were produced, for which the counting rate for spontaneous fission was 1-2 counts per day, which corresponds to a $3 \times 10^{-11}\text{g/g}$ concentration of the nuclide sought for. The results of this investigation, together with other data obtained by us earlier (Ref.36,43-45,70-72,75,76) are shown in Fig.6, where the specific spontaneous fission activity of the samples is presented as a function of the uranium concentration. The attempts at a further concentration of this activity in the samples of a still smaller weight appeared to be unsuccessful due to losses that increased at each stage of processing.

Therefore, in our subsequent experiments we aimed at separating a much greater quantity of the element sought for from the initial Cheleken brine. Such extraction requires the processing of great quantities of brine since the concentration of this element in it was very low: according to estimates which followed from the saturated resin data, the counting rate for spontaneous fission events for the Cheleken water was only 1-2 fissions a day per ton. Bearing in mind the technological difficulties of the chemical processing of massive samples, priority was given to such chemical methods which could allow us to process 10-100 tons of brine at one stage by concentrating the nuclide sought in samples several kilograms in weight (Note b). Different methods were used to carry out such extraction as concentration on a metal, ion-exchange concentration, co-precipitation with hydroxides and sulfides, and extraction into the organic phase. The complexity of this work was determined by such factors as the necessity to maintain the constant, in time and in the whole volume of the brine, values of acidity and redox potential, by the presence of organic substances in the water, the inconsistency of its general salt and microelement contents. The essential difficulty of the task was probably associated with passing from a factor of 100 concentration achieved earlier (Ref.45) to that by a factor of 10^3-10^4 . In fact, one cannot exclude the high stability of the lower valent states (e.g., 2,1 or 0) of the element sought for, the high solubility of its sulfides or the amphoteric character of hydroxides, which could lead to a very narrow, if not zero, range of the conditions required to obtain such a factor of concentration.

So far, we have not achieved the desired factor of concentration, although all the above methods enabled us to obtain samples 1-3 kg in weight, which display the spontaneous fission activity of the nuclide under study with a counting rate of 0.5-1 per day. Such reproducibility of the results by using different chemical methods indicates that the observed activity has not been produced by some random admixture of the pure ^{252}Cf isotope, and stimulates further investigations. However, it is clear that new approaches are needed to solve this problem.

One of these approaches is possibly associated with the search for such terrestrial samples, in which the initial concentration of SHE is much higher than in Cheleken hot spring waters.

Note a. This work was done by Yu.S.Korotkin.

Note b. This work was carried out by V.Ya.Vyrovaev et al.

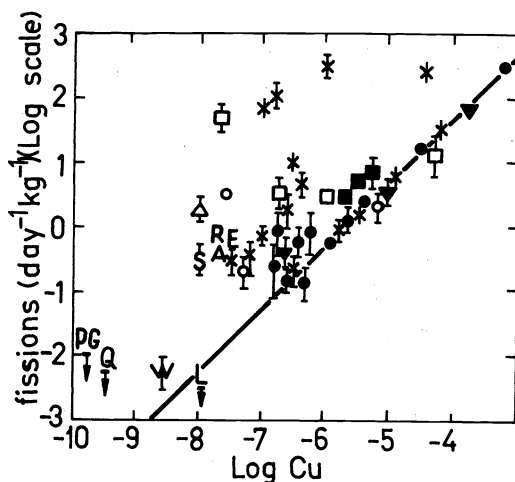


Fig.6. Specific spontaneous fission activity of samples vs uranium concentration. The full line corresponds to the counting rate due to the spontaneous fission of uranium; S,A, and E are Saratov, Allende and Efremovka, respectively; R - saturated resin, PG - Plexiglass, Q - melted quartz, W - Cheleken water, L - metallic lead. * - the products extracted from Cheleken hot springs; \bullet \circ - Pb-Zn ores and products of their processing; \blacksquare \square - Fe-Mn nodules and products of their processing; Δ - thermal sublimation of Allende; \blacktriangledown - other samples.

In this connection it is of interest to expose the possible correlations between the concentration of SHE and other chemical elements. We noted earlier (Ref.45) that the spontaneous fission activity detected in meteorites and Cheleken hot springs did not show any pronounced direct correlation with any of the known elements. In fact, in quite a number of investigations on the search for SHE, using the method based on detecting spontaneous fission (Ref.36, 38,39,49, 50,53,58-63) negative results have been obtained for numerous samples enriched in platinum metals, gold, volatile metals (Hg, Tl, Pb, Bi), rare earths and other lighter elements. Therefore the sensitivity required to detect the spontaneous fission activity of various known elements was much higher in the above experiments than in our measurements with meteorites or geothermal waters. Fireman et al. (62) also point out this fact but see some contradiction in it.

However, in our view, this indicates that the spontaneously fissioning nuclide detected in meteorites and in the Cheleken water cannot be assigned to any isotope of a known element which displays unusual properties due to isomerism or weak spontaneous fission branch. On the contrary, if this nuclide actually belongs to the SHE region, it will rather correlate with its lighter homolog in the case of a not very high concentration of the latter (e.g., 10^{-5} - 10^{-3} g/g) and not follow it in the course of the further concentration. Consequently, there can exist such terrestrial samples, in which the spontaneously fissioning nuclide sought is present in a proportion much higher than in meteorites which represent the averaged composition of the Solar System substance. It is not excluded that such samples may be abyssal ferro-manganese nodules or polymetallic lead-zinc ores, some data on which are presented in Fig.6. The further searches for SHE in similar samples and the establishment of their possible correlations with the contents of small admixtures of known elements seem to be of interest.

Another approach is associated with the development of new sensitive methods for the search and identification of SHE. This necessity is dictated by the fact that all the previously used methods have disadvantages which hinder or eliminate their application in the study of a great number of samples or in the process of successive chemical concentration. Thus, for example, in the search for the spontaneous fission of SHE one has to restrict the range of the samples to be studied to those in which the contents of uranium is lower than that of the Earth's crust (3×10^{-6} g/g), since at higher concentrations its spontaneous fission hinders the detection of other emitters. On the other hand, the heavy chemical elements that are prone to the manifestation of more than one valent state are often concentrated in natural samples enriched in uranium. It is not excluded that such behaviour is also characteristic of the SHE sought for. In this case the search for a SHE by detecting its spontaneous fission would eliminate the possibility of studying the most interesting samples. A disadvantage of this method is also the fact that to achieve a high sensitivity one has to conduct measurements with samples of great weight (10-20 kg), and therefore, tons of initial raw material are to be processed in the course of chemical concentration.

Other methods used do not provide the sensitivity required to detect SHE in natural samples. Therefore, they should be combined with the preliminary chemical concentration. For example, in the case of observation of characteristic X-rays induced by synchrotron radiation (Ref.82,83), the required concentration factor reaches 10^5 - 10^6 relative to the substance of the Allende meteorite.

In an attempt to find the shortest routes for the identification of the spontaneously fissioning nuclide detected in meteorites and Cheleken hot springs, we performed two experiments, each of which, in the positive case, could give a definite solution to the whole problem. In one of these experiments (Ref.84) the samples ranging from several grams to several dozens of grams in weight, in which spontaneous fission was observed at a counting rate of about 0.25 per day, were heated successively in hydrogen and oxygen flows, and the chemical elements that are volatile in the temperature range 20 to 700°C were separated from them. The obtained fractions were irradiated with an α -particle beam to observe the induced fission of SHE, characterized by the total kinetic energy of fragments of ≤ 200 MeV. Such fission fragments have not been detected in these experiments. Unfortunately, this results did not allow us to draw any definite conclusions since it could be explained equally either by the presence of a very small amount of SHE ($<10^9$ atoms) in the initial samples, or by very large losses of SHE (by a factor of >100) during the thermochromatographic treatment, which required an increase in a factor of 10^6 - 10^7 concentration of this element.

In the other experiment, we irradiated the samples under study with resonant neutrons, counting on that the initial long-lived nuclei of a SHE could be transformed, as a result of neutron capture, to new nuclei with short spontaneous fission lifetime. The counting rate of spontaneous fission in the samples could thus be increased considerably, which much simplified the establishment of its nature. However, the irradiation of the fractions obtained from the Cheleken water with an integrated flux of resonant neutrons of 10^{21} cm $^{-2}$ (the flux of thermal neutrons was 10^{22} cm $^{-2}$ in this case) did not result in a noticeable increase of the counting rate against the background of the spontaneous fission of ^{244}Cm which could have been formed as a result of neutron capture by the nuclei of the ^{238}U admixture. Even if there was some increase, it did not exceed a factor of 100. Such a slight increase can be explained assuming that the value of the resonance integral of the neutron capture of long-lived SHE nuclei is <100 barn, and the decrease in half-life does not exceed 10^3 . Such an assumption is quite admissible and therefore it is difficult, on the basis of this experiment, to draw any conclusions neither supporting nor opposing the hypothesis about SHE.

The negative results obtained in the two experiments described above, indicate the necessity of the especially thorough performance of the further experiments and suggest that the solution of the above problem demands the use of a more sophisticated method. We believe that the mass-spectrometric method is a promising one in this respect. The present-day development of mass-spectrometry permits detection of small admixtures of chemical elements in different samples, at a level of 10^{-7} - $10^{-8}\%$ of atoms. The further increase in sensitivity requires eliminating the background from the scattered ions of the sample matrices, as well as that due to charged molecules and radicals, which initiate the mass lines with $A \geq 238$. These sources of background were reduced sharply in the ultrasensitive systems designed for identification of particles (Ref.85,91) and using electrostatic accelerators. However, the application of such a system in the search for SHE enabled one to achieve the sensitivity of only $10^{-8}\%$ of atoms. On the other hand, the elimination of the background from scattered ions with strong mass lines is possible within the framework of classical mass-spectrometry, if one uses the multi-stage separation by means of tandem mass-spectrometers (Ref.86). However, the application of such tandem systems requires the exact knowledge of the mass of the isotope sought for. In addition, the adjustment of such setups is a very complicated task.

With all these factors in mind, we are now designing a mass-spectrometer which would permit the multiple isolation of SHE ions in the chosen range of mass numbers and the suppression of the background due to charged molecules and radicals by means of a gas target placed in the ion beam. A schematic diagram of this mass-spectrometer is presented in Fig.7. It includes a laser ion source (Ref.87), which provides up to 10^{10} ions per one laser pulse through a slit. The consumption of the substance of the sample under study is smaller than $1 \mu\text{g}$ per pulse in this case. The ions accelerated to an energy of 100 keV pass through a gas target to the entrance of an achromatic ion-optic system, which includes two dipole and several quadrupole magnets. Such a system can be adjusted for the transmission of ions with masses lying in the required range, and it gives the image of the source slit, which serves as an object for the next stage - the dipole magnet separating in mass the ions that have passed through the achromatic system. The registration of ions is performed with channel plates during the time intervals, when the ions of the required mass are expected to arrive at the focal plane. This provides an additional decontamination of the ions. The mass resolution is equal to about 500 taking into account the energy spread of the ions emitted by the laser source and ion scattering in the gas target. Such a resolution is, however, sufficient for the reliable isolation of SHE at concentrations of 10^{-11} (and possibly $10^{-12}\%$) of atoms. To achieve such sensitivity would require the consumption of samples weighing up to several dozens of mg, while the duration of one analysis would lie between dozens of minutes and 2-3 hours.

It would be desirable that other groups engaged in the SHE problem take part in the development of mass-spectrometers, that are necessary for carrying out the search for these elements in nature. In particular, not all the possibilities of mass-spectrometry using tandem generators have apparently been exhausted.

In conclusion, to summarize the results of 12 years studies on the search for SHE in nature, performed in Dubna, we would like to note the following.

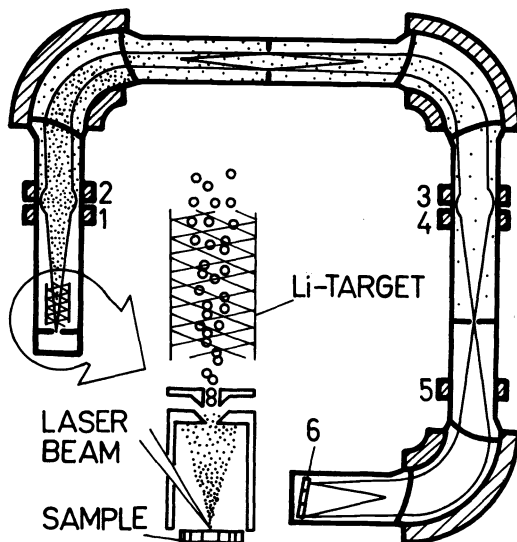


Fig.7. Schematic drawing of the mass spectrometer: (1-5) - quadrupoles, 6 - channel plates.

The method of observing atomic nuclei in galactic cosmic rays by revealing the tracks accumulated in meteorite minerals, has been developed to be practically fully reliable. The rate of scanning meteorite olivines has been achieved to provide a high sensitivity of the experiments. In the measurements performed within a short period of time, about 400 tracks of thorium-uranium nuclei have been detected. Two tracks have been revealed, the length of which corresponds to atomic nuclei with $Z \approx 110$. The volume of the minerals examined can be increased rather easily by a factor of 10, which should lead to more definite conclusions about the possibility of the existence of SHE nuclei in galactic cosmic rays.

The methods for counting the rare events of spontaneous fission, developed at Dubna, have enabled one to achieve a record sensitivity in searches for SHE and to study the largest set of natural ores and minerals. As a result, a new natural spontaneously fissioning nuclide has been detected in the samples of carbonaceous chondrites and in the geothermal waters of the Cheleken Peninsula; this nuclide can be assigned to the region of superheavy elements with high probability. To extract this nuclide from natural samples and to concentrate it, different chemical methods have been used, which have enabled one to prepare samples up to several grams in weight, with a counting rate for spontaneous fission of several decays per day. The attempts to obtain samples with a counting rate of 10-100 decays per day in the course of chemical extraction from the Cheleken water, have not yet produced the desired result. This is obvious due to the non-trivial character of the problem, associated with the absence of any information on the chemical behaviour of the element sought and with the difficulties arising during the processing of great amounts of natural brines.

The experiments, which could be crucial for the identification of the nuclide detected in case of their positive result (the observation of the induced fission by an α -particle beam and irradiation with high fluxes of resonant neutrons) have not yielded any definite results. It makes us approach the performance of further experiments very carefully and indicates the necessity to use new, more sensitive and complicated methods.

The further studies should include the development of chemical methods for the extraction of SHE, the search for natural samples with a concentration of the element sought for higher than that in meteorites and hot spring waters, the development of new physical methods for the detection and identification of SHE, of which mass-spectrometry seems to us to be the most promising technique. In case the mass-spectrometric methods are developed to the level sufficient for solving this problem, the application of neutron multiplicity detectors in the search for SHE will be somewhat reduced. However, we believe that similar detectors may be very useful in the search for the nucleon decay phenomenon. This problem has become topical in connection with the predictions of the unified theories of weak, electromagnetic and strong interactions (see Ref.92). As estimates show, one can observe nucleon decay with neutron multiplicity detectors having a sensitivity corresponding to the half-life limit of 10^{31} years. Since such measurements depend to the least degree on the assumption about the expected modes of nucleon decay, their performance is of great interest.

Many chemists and physicists have been dreaming for years of revealing in nature chemical elements heavier than uranium. Rather often this dream seemed to be realizable and promised to open new horizons in the Sea of unstable heavy elements. Still more often it brought the disappointment of the hopes that did not come true. Nevertheless it is difficult for us to abandon this dream. Especially now when some results obtained in Dubna give more evidence for

the possible existence of a relict SHE. This inspires us to make new and new efforts to obtain the final and, as we hope, positive solution of a most interesting problem.

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