Natural superheavy nuclei in astrophysical data

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Abstract. The paper presents the summary data of the authors’ research within the framework of the OLIMPIYA project (the Russian acronym of OLiviny iz Meteoritov — Poisk tyazholykh i sverkhtyazholykh Yad / Olivines from meteorites: Search for heavy and superheavy nuclei) and results of track analysis for heavy cosmic ray nuclei ($Z = 26–129$) in olivine crystals from meteorites using an original processing technique. A total of 21,743 tracks of nuclei heavier than iron have been identified in meteoritic matter to date to form the largest database within this charge range. The database includes three tracks of superheavy nuclei with the lifetimes of about a few decades, which can be considered as direct experimental evidence for the existence of natural superheavy nuclei from the “island of stability”. Comprehensive comparative analysis of data from two meteorites with different cosmic ray exposure ages, Marjalahti (from 178 to 205 Myr) and Eagle Station (from 35 to 71 Myr), is presented for the first time. The results are discussed within the existing concepts of nuclei formation in astrophysical processes.

Keywords: superheavy nuclei, galactic cosmic ray, olivine, image recognition
1 Introduction

The paper presents a summary of the authors’ work on the use of meteoritic olivines as effective detectors of extraterrestrial nuclei. Emphasis is made on the search for heavy and superheavy nuclei in galactic cosmic rays and the analysis of possible ways of their formation and propagation in the Universe. The work is directly related to the fundamental problem of the limits of the Periodic Table of Elements, the 150th anniversary of which is celebrated in 2019 by the UNESCO decision.

Although the standard electrodynamics allows for nuclei with atomic numbers greater than 170 [1], the only massive natural stable chemical element on Earth is uranium with a nucleus charge of 92; all nuclei heavier than that have been obtained artificially. Instability of nuclei heavier than uranium results from a faster increase of Coulomb repulsion of protons in comparison with attraction caused by nuclear forces when the number of nucleons in a nucleus increases. The nuclear shell model [2] predicts “islands of stability” for superheavy nuclei. According to theory, the arrangement of nucleons into complete shells within the atomic nucleus creates states of the largest binding energy, which for the so called “magic” numbers of neutrons and protons (2, 8, 20, 50, 82, 126 — calcium, tin, lead, etc.) significantly increase the height of the nuclear fission barrier [1, 3, 4]. The half-lives of the stable isotopes of these elements can be from several minutes to several years (up to thousands of years, according to over-optimistic estimates [5]). Attempts to produce new superheavy elements and to synthesize elements with \( Z \geq 100 \) continue on accelerators [6–8].

Synthesis of superheavy elements poses certain experimental problems. All elements with \( Z \geq 100 \) have been produced on high-power accelerators in heavy ion-induced fusion reactions. The probability of these reactions to occur and their products to survive fission is extremely low due to the necessity to create extremely high energy densities and high neutron fluxes, so accelerator experts try to make a proper choice of target projectile and projectile’s energy combinations. The time required to synthesize one atom of elements 119 or 120 is estimated to be hundreds of days [9]. The challenge is aggravated by the fact that laboratory-produced superheavy elements are represented by neutron-poor isotopes, which are generally unstable [10]. The resulting dominance of Coulomb repulsion leads to a spatial heterogeneity and non-sphericity of the nuclei, thus stimulating their decay.

If theory is right, and the “island of stability” does exist, it should be possible to detect these nuclei in nature. Astrophysical studies are recognized as a promising way to search for these nuclei [11, 12]. As the fraction of heavy element nuclei in cosmic rays is very small and does not exceed 0.5%, their flux in near-Earth space is about a mere 1–2 nuclei/m\(^2\) per year. This results in an altogether small number of nuclei from the region of transuranium
elements with single events related to nuclei with $Z > 92$ registered in direct balloon [13–15] and satellite [16–19] experiments with cosmic rays. This limitation can to a significant extent be overcome by using natural track detectors, which are meteorites and lunar samples [20, 21]. Estimates by Prof. G.N. Flerov show [22] that the study of one gram of mineral from a meteorite with a cosmic ray exposure (CRE) age of $\sim 10^8$ years is equivalent to an experiment for the direct irradiation of a ton of nuclear emulsion for one year on the Earth orbit.

Study of olivine crystals from the Marjalahti and Eagle Station pallasites for direct registration of tracks of heavy nuclei was initiated in the 1970s at the Flerov Laboratory of Nuclear Reactions, Joint Institute for Nuclear Research (Dubna) [23]. These experiments resulted in fixation of several super-long tracks of nuclei heavier than uranium [24]. In 2005, two fragments of these meteorites were transferred to the Lebedev Physical Institute of the Russian Academy of Sciences to carry out further studies using new techniques and apparatus [25]. In subsequent years, this research, being carried out together with colleagues from the Vernadsky Institute of Geochemistry and Analytical Chemistry, Russian Academy of Sciences, provided for accumulation of a large database of heavy and superheavy GCR nucleus tracks in meteoritic olivine [26, 27]. Analysis of these data confirmed the presence of tracks of transuranium and transfermium elements in meteoritic matter. Our paper presents the summary results of the research within the framework of the OLIMPIYA project to search for and identify heavy and superheavy nuclei of galactic cosmic rays (GCR) and confirm experimentally the existence of transuranium and transfermium elements in nature.

Two results presented in the paper motivated its writing. First, we obtained new results of track processing, which considerably increase the database statistics. Second, we compared data from two meteorites of different radiation ages, Marjalahti and Eagle Station, to enable an experimental database to be set up to test models of the formation of heavy and superheavy elements in various sources and to identify the main mechanisms of their appearance as the GCR fraction.

2 Olivine crystals as natural track detectors

The use of stony-iron meteorites as natural detectors for the search for superheavy nuclei in nature, namely in galactic cosmic rays, seems promising for a number of reasons. First, the exposure times of meteorites are millions of years [28], which increases the probability of fixing superheavy nuclei, the fraction of which in cosmic rays is negligible. Second, since meteorites travel far beyond the Solar System, they can capture superheavy nuclei in relative proximity to the source of their formation, in contrast to near-Earth detectors. And, in addition to stable nuclei, meteorites are capable of fixing unstable isotopes of superheavy nuclei with lifetimes longer than their times of flight from the source to meteorite.

The presented research makes use of natural track detectors, which are translucent crystals of magnesium-iron silicate olivine ($\text{Mg}_{0.8}\text{Fe}_{0.2}\text{SiO}_4$) extracted from Marjalahti and Eagle Station pallasites and prepared for microscopic measurements. Contrary to other types of track detectors (photoemulsion, plastic), olivine has no background tracks from nuclei lighter than iron, which is due to a rather high threshold of specific energy losses of the charged projectile required to form its chemically etched track in olivine (the threshold value of ionization losses of energy for olivine is about $18 \text{ MeV}/(\text{mg} \cdot \text{cm}^2)$ [29]).

Research into tracks of GCR nuclei in olivines from pallasites is based on an original method of controlled etching [26, 30] exploiting the fact that areas of a crystal damaged by a passing charged particle have a higher chemical activity compared to intact areas. The
dissolution of material by a multicomponent chemical etchant proceeds at a much higher rate in this area to enable the “development” of nuclear tracks making them visible in translucent olivine under an optical microscope and available for analysis. Identification of cosmic ray nuclei in olivine crystals is based on the study of the dynamic and geometric characteristics (etching rate $V$ and residual path length $L$) of chemically etched segments of the nuclei deceleration trajectories before the nuclei are completely stopped in olivine crystals. These characteristics depend on the extent of crystal lattice damage and on ionization caused by the passage of a swift heavy ion and, therefore, on the magnitude of its charge.

The polished surface of olivine crystals is subjected to multistep chemical etching and processing; the parameters of tracks visible at a given treatment step are measured. After the measurements, a 50–100 $\mu$m crystal layer (accurate to $\mu$m) is removed, and the procedure is repeated. The new detected tracks are being linked with those in the previous layer. The complete track lengths are summed up with account for the trajectory slope to the crystal surface and the thickness of the removed layer. Such successive operations of polishing/etching/search/track registration enable the fixation of a) the spatial orientation of tracks in a crystal, b) the nucleus stopping point or, which is the same, residual range $L$ of a projectile and c) the etching rate $V$ at various segments of the nucleus trajectory.

Search for and analysis of tracks is performed on the PAVICOM automated measuring setup [31]. Figure 1 presents micrographs (with $60 \times$ magnification) of typical etched nucleus tracks in the developed olivine crystals. Reflecting the dependence of the energy loss on the energy of a projectile, the tracks are formed of two parts: cylindrical, at the end of the trajectory; and needle-like, at the trajectory segment corresponding to larger projectile energies. Figure 1(a) clearly shows the boundary separating the track into two segments, the channel diameters of which differ severalfold; in figure 1(b), this transition is less pronounced.

![Figure 1](attachment:image.png)

**Figure 1.** Micrographs of GCR superheavy-element nuclear tracks registered in olivine crystals from pallasites. Field of view $150 \times 80 \mu$m (a) and $100 \times 55 \mu$m (b).

Irradiation of olivine crystals with different swift heavy ions on accelerators of IMP (Lanzhou, China) and GSI (Darmstadt, Germany) [30], as well as comparison with the results of simulations based on an original numerical model of track formation and etching in olivine [32], allowed for charge calibration of the data obtained in the OLIMPIYA experiment. These studies and subsequent analysis gave us an opportunity to study in detail the mechanisms involved in the formation of detected tracks as well as to obtain the dependence of the etching rate on the residual range and nucleus charge $V(L,Z)$, which provided for the determination of charges of the registered nuclei to an accuracy of $\pm 1$ to $\pm 2$ units for $Z < 92$. Within the range of $67 < Z < 92$, the dependence was fitted by a five-parameter function...
allowing for its extrapolation to larger $Z$ values [26].

A large database of the processed tracks of GCR nuclei in olivines from Marjalahti and Eagle Station pallasite meteorites has been accumulated over about a decade of the experiment. Our results are widely used in discussions on the origin of galactic cosmic rays and on the hypothesis of the existence of the “island of stability” nuclei [9, 12, 33–36].

3 Results and discussion

According to the current views on the origin of heavy and superheavy nuclei in the Universe, elements heavier than iron can be formed by two scenarios — slow or rapid neutron-capture processes (s-processes and r-processes, respectively) [37, 38]. As the result of both these processes, the nucleus rich in neutrons experiences a $\beta$-decay that converts neutrons into protons and increases the nucleus charge. In an s-process, production of nuclei is based on the gradual capture of neutrons with subsequent $\beta$-decays, i.e. the process moves along the chain of re-emerging short-lived proton-enriched nuclei. The process is “slow” because it needs sufficient time for the realization of the radioactive decay before the next neutron will be captured by a nucleus. The s-process is realised at “low” neutron fluxes/densities ($10^{5}$ to $10^{11}$ cm$^{-2}$s$^{-1}$) and is not able to produce heavy elements adjacent to uranium.

At a neutron density of $10^{10}$ n/m$^3$, the transformation of Fe to Pb takes place. The s-process responsible for the formation of elements up to $Z = 83$ can occur, in particular, in the Red Giants. Nuclei in the range of $Z = 84–89$ do not have stable isotopes, that is why it is impossible to “fill” this segment of GCR charge spectrum with s-process products. On the other hand, nuclei with $Z = 90$ (thorium) and $Z = 92$ (uranium isotopes $^{235}$U and $^{238}$U) do exist in nature. This can be explained by the assumption that they are formed by the consecutive capture of several neutrons by unstable nuclei in the r-process.

The r-process is a successive neutron capture occurring faster than the $\beta$-decay does. The heaviest nuclei are formed in a “strong” (or “violent”) r-process at neutron densities higher than $10^{19}$ cm$^{-3}$ (and reaching $10^{24}$ or even $10^{27}$ cm$^{-3}$), when the nucleus of iron captures 100 neutrons and more. According to different theoretical scenarios, conditions for superheavy nuclei formation can be realized in supernovae (SNs) [39] or in neutron stars or else neutron star-black hole mergers (the latter two both indicated as NSMs) [40, 41]. NSMs are considered to be the most powerful sources of r-process matter, ejecting on average from 100 to 1000 times more r-process material than SNs do. Therefore, although the NSMs (manifesting themselves as macronovae or kilonovae) occur significantly more rarely than core-collapse supernovae (the frequency of SN collapses in the Milky Way Galaxy is $\sim 1/30$ y$^{-1}$, i.e., several orders of magnitude higher than the frequency of NSM events, which is $10^{-5}$ y$^{-1}$), they could potentially be the dominant mode of producing heavy elements [33, 42].

Synthesis of superheavy elements in kilonovae is confirmed by recent data on the electromagnetic spectrum of the event GW170817 interpreted as a signal from an NSM, in which two components of the emission are distinguished; one of them consists mainly of light (atomic mass number less than 140), the other of heavy elements of the r-process (atomic mass number more than 140) [43]. These observations became available through the discovery of gravitational waves from inspiralling neutron stars in the LIGO-Virgo experiments [44], confirming and significantly improving the sky localization of this event.

Because of a large difference between the exposure times of meteorites and artificial orbital satellites, they must register nuclei synthesized in different sources. The satellites are exposed to current fluxes of nuclei in near-Earth space and register mainly fragments of nuclei.
formed in neutron matter fusion in relatively frequent SN explosions. In close vicinity to the Solar System (1.5–3 kpc), i.e. at distances from which the formed nuclei keeping information about details of their birth come, only several NSM events occur in 100 million years, which means that the exposure times of orbital satellites are too small to fix nuclei from NSMs. Meteorites can fix bursts of fluxes of superheavy nuclei born in NSMs in the $r$-process of neutron matter fusion, in relative vicinity to which they occurred when exposed to radiation formed. In this way, meteorites hit by superheavy element ejecta from NSMs become witness of cosmic events that had occurred millions of years ago hundreds of light years away from the Earth.

This hypothesis is substantiated in [33], based in particular on the data obtained by the OLIMPIYA, Ariel-6 [17], HEAO-3 [18], UHCRE [19] and SuperTIGER [45] experiments. The authors of [33] performed model calculations for the evolution of the energy spectrum of superheavy nuclei formed in an NSM $r$-process for various models of acceleration of particles in space. The analysis given in this work has shown that the OLIMPIYA data are in good agreement with predictions of the NSM model.

Since our last comprehensive review [26], we have been able to significantly improve the reliability of the experimental results, almost doubling the statistics of the measured tracks. The statistics accumulated during this period amounted to 21,743 processed tracks of heavy and superheavy nuclei with charges from 26 (the threshold value) to, at least, 119. Figure 2 presents the renewed data of the relative distributions of the nuclei recorded in both meteorites with 95% reliability error bars. (In the next figures all the data are given without the error bars in order to facilitate the required comparisons.) The data in figures 2, 3, 4 are normalized so that the sum of all probabilities over all integer charges gives unity. As in [26], all our results are normalized to the abundance of iron nuclei $A_{(26\text{Fe})} = 10^6$.

Table 1 and figure 3 demonstrate the difference between our updated data and the results of the satellite experiments [17–19]. Comparison of the nuclei abundance ratios demonstrates that the fractions of superheavy nuclei registered in the meteorites significantly (up to 100%) exceed those registered by satellite instruments on the Earth orbit, i.e. indicates that these nuclei were apparently born in $r$-processes that took place in very rare NSM events. Table 1
and figure 4 also show the difference between the relative abundances of nuclei registered in Marjalahti and Eagle Station.

The number of nuclei that passed through meteoritic matter depends on the CRE age of a meteorite. The estimated exposure times of the meteorites used in our research range from 35 to 71 Myr for Eagle Station and from 178 to 205 Myr for Marjalahti [46]. These times are much larger than 17 Myr which, according to estimations made in [33], are required to fix superheavy nuclei by a meteorite. Due to a considerable difference between their CRE ages, these two meteorites have witnessed, with high probability, a different number of NSM events. Citing [33], “if the time averaged fluxes measured by meteorites with different ages show different values, it will be a smoking gun of the NSM contribution of UHCRs” (ultra-heavy cosmic rays). Now we can conclude that the difference between the abundance ratios of superheavy nuclei registered in Eagle Station and Marjalahti (see Table 1) looks like a direct indication of this smoking gun of proof.

As for the data of two meteorites, Figure 4 shows that the probability of registering nuclei with $50 < Z < 60$ for Eagle Station is higher than for Marjalahti; for $Z > 65$, it is the

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**Table 1.** Abundance ratios based on the satellite experiments [17–19] and the OLIMPIYA updated data.

|                  | HEAO-3          | Ariel-6         | UHCRE          | OLIMPIYA Marjalahti | OLIMPIYA Eagle Station |
|------------------|-----------------|-----------------|----------------|---------------------|------------------------|
|                  | (Binns 1985, 1989) | (Fowler 1984, 1987) | (Donnelly 2012) | (Marjalahti)        | (Eagle Station)        |
| Actinides/Pt     | $0.0241^{+0.022}_{-0.010}$ | —                | $0.025 \pm 0.005$ | $0.05 \pm 0.008$ | $0.038 \pm 0.008$     |
| Actinides/Subactinides | $0.0186^{+0.018}_{-0.010}$ | 0.04             | $0.020 \pm 0.004$ | $0.036 \pm 0.006$ | $0.028 \pm 0.006$     |
| Pb/Pt            | $0.30 \pm 0.081$ | $0.40 \pm 0.10$ | $0.25 \pm 0.04$ | $0.23 \pm 0.026$ | $0.24 \pm 0.03$       |
| HS*/PbPt         | $0.16 \pm 0.06$ | $0.27 \pm 0.07$ | $0.19 \pm 0.03$ | $0.64 \pm 0.03$   | $0.52 \pm 0.035$      |
| LS**/PbPt        | $1.1 \pm 0.2$  | 1.45$ \pm 0.25$ | —              | 1.98$ \pm 0.065$ | 2.1$ \pm 0.13$        |

* Heavy Secondary ($70 \leq Z \leq 73$).
** Light Secondary ($62 \leq Z \leq 69$).
other way round. Thus, the average slope of the charge dependence decreases with exposure time and could be explained by the low abundance of the heaviest components in cosmic rays. This fact is also manifested in the abundance of transuranium nuclei (see Table 1); it is much higher in Marjalahti than in Eagle Station, which could be expected due to a longer exposure time of the former meteorite. The observed difference in the abundance of heavy nuclei can also be explained by different distances from the meteorites to the NSM during their irradiation.

The experimentally recorded difference between the relative abundances of nuclei registered in meteorites with different exposure times, as well as the difference between the meteorite data and the results of the satellite experiments, can be used to test models of nuclei propagation to the Solar System, including fragmentation effects of heavy elements generated by NSMs.

The most significant result of the OLIMPIYA experiment is the detection and identification of three transfermium nuclei with charges estimated as $119_{-6}^{+10}$, which can be considered as the first direct evidence of the existence of naturally occurring stable superheavy nuclei. In 2013, the OLIMPIYA group reported the detection of three tracks of particles with atomic numbers in the interval from 105 up to 130 [30]. The track lengths corresponding to the registered superheavy nuclei exceeded 500 $\mu$m, and the etching rates $V$ were greater than 35 $\mu$m/h [26]. Taking into account that the experimentally measured maximum rate of track etching for uranium nuclei in olivines before their stoppage is $26\pm1$ $\mu$m/h, it becomes clear that the charges of these nuclei significantly exceed the charge of uranium. The charge assessment for these nuclei was based on the dependence of the etching rate near the stoppage point on the charge value (see Section 2). The dependence of the etching rate on the charge was approximated by a straight line along the available five experimental points up to the value of $Z = 92$. Further extrapolation of this straight line up to the etching rate value of 35 $\mu$m/h and the regression analysis performed made possible the assessment of charge of these nuclei with 95% probability.

In the context of the above, it can be argued with a high degree of certainty that the data obtained in the OLIMPIYA experiment supply arguments supporting the existence of...
the theoretically predicted “island of stability” of long-lived transfermium nuclei in nature. The three ultraheavy nuclei detected can be considered as experimental confirmation of the existence of transfermium elements in nature and represent a result of high significance.

Of great interest are the chemical properties of the elements closest to $^{118}\text{Og}$ with larger nucleus charges, not yet obtained on accelerators. The peculiarities of the electronic structure of atoms with superheavy nuclei are determined by relativistic effects and may differ sharply from the “usual” electronic properties of atoms with less heavy nuclei. Since $^{118}\text{Og}$ is an inert gas (or liquid) and completes a period of the Periodic System of Chemical Elements, elements 119 and 120 should belong to the 8s-block and have the properties of alkaline and alkaline-earth “metals”, respectively. But substances composed of superheavy atoms may exhibit still unknown and unusual chemical and physical properties. Prediction and registration of these properties pose a fundamental problem.

4 Conclusion

The presented material demonstrates the efficiency of the meteorite method for studying the heavy component of galactic cosmic rays. To date, 21,743 tracks of ions heavier than iron have been detected and identified in the OLIMPIYA experiment, providing statistically significant data on the relative abundance of heavy and superheavy nuclei in galactic cosmic rays. These data are compared with the results obtained in satellite experiments, as well as in two meteorites with different ages of exposure to cosmic rays. The results have been obtained and are discussed within the framework of the existing concepts of the formation of heavy and superheavy nuclei in astrophysical processes in the Galaxy. The difference between nuclei abundances in two meteorites is a strong evidence of the superheavy nuclei formation in mergers of neutron stars (or neutron star-black hole mergers) and support the hypothesis that the condition for their synthesis is a huge free neutron density granting a rapid capture of neutrons and suppressing $\beta$-decay.

One of the most important results of the experiment, which is the registration of three tracks of nuclei with charges in the range of $113 < Z < 129$, has been confirmed.

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