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V.P.Perelygin, S.G.Stetsenko, H.J.Crawford*,
T.J.M.Symons*

ANALYSIS OF THE RESULTS OF CALIBRATING METEORITIC OLIVINE CRYSTALS WITH ^{238}U NUCLEI AT THE BEVALAC ACCELERATOR

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*Lawrence Berkeley Laboratory, Berkeley,
California 94720, USA

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The present experiments on the sensitivity calibration of meteoritic olivine crystals at the external ^{238}U beam from the Bevalac facility (LBL, Berkeley) were initiated with the aim to clarify the origin of the longest groups of tracks due to Galactic cosmic ray nuclei, which were measured in these crystals at the LNR, JINR during 1980-1987 /1,2/.

It is necessary to point out that the study of the most heavy component of cosmic-ray nuclei was initiated by Fowler et al. /3/ in 1967, and then performed by a number of research groups /4,5/ using the stacks of nuclear emulsions and plastics, exposed on balloons and on the orbital station Scylab.

From the very beginning of the 80's, two systems of big electronic detectors started to register cosmic-ray nuclei at the orbital station "ARIEL-6" and "HEAO-3" /6,7/. Due to a rather low collecting power only a few Th-U cosmic-ray nuclei have been definitely recorded in all of these experiments.

The data presented in /6,7/ shows that the abundance of cosmic-ray nuclei with $Z \geqslant 90$ is $(1-3)x10^{-7}$ relative to Fe group nuclei and does not exceed 1-2 $nucl/m^{-2}$ per year for the region of energies $E \geqslant 1.3$ GeV/nucl.

Such rather low intensity handicaps very much both investigations of Th-U nuclei and especially the search for the $Z \geqslant 110$ nuclei in the experiments on the direct registration of such nuclei in outer space.

The other way of galactic cosmic ray investigations was based on the ability of a silicate meteoritic crystals-pyroxenes, olivine, feldspar,etc. to register and to store for many tens and hundreds M.Y. the tracks due to nuclei with $Z \geqslant 20$ (the Fe group and heavier nuclei).

But obtaining quantitative information on the charge and energy spectra of ultra heavy cosmic-ray nuclei based on a fossil track study in extraterrestrial crystals meets a number of methodical problems, such as a very high background due to Fe group nuclei, 10^{10}-10^{11} tracks cm^{-3}, which excludes the possibility of revealing the full length of tracks from $Z \geqslant 50$ nuclei /9/. Another problem is connected with partial annealing of the track in silicate crystals under the outer space conditions, which does not allow comparison of the etchable track length of fossil tracks and "fresh" tracks - due to calibration of the same crystals by accelerated heavy ions with a known Z.

Because of the above difficulties several attempts of some groups of authors, at the end of the 60's-beginning of the 70's to investigate the most heavy component of galactic cosmic-ray nuclei by the track study in meteoritic and Moon crystals yielded only qualitative results in the region of cosmic-ray nuclei with $Z \leqslant 83$ /9-12/.

At the first stage of the investigations, performed in the LNR of JINR during 1973-1979 large transparent meteoritic olivine crystals from pallasites were chosen as an object of the study of galactic cosmic nuclei tracks. As is quoted in the book of Chirvinsky /13/ the olivine crystals in each given pallasite are very homogeneous in chemical composition. In such a pallasite olivine minerals form monocrystals up to 1-2 cm in diameter; it occupies 40-50% of the volume of this kind of meteorites.

The detailed measurements of the track density due to Fe group nuclei performed in crystals from 16 pallasites led to the choice of the pallasites Marjalahti and Eagle Station as most suitable objects for further studies.

The track density of Fe group nuclei in olivine crystals from some locations of these two meteorites reach 10^{10}-10^{11} tracks per cm^3, and 10^6-10^7 $tr.cm^3$ due to $Z \geqslant 36$ nuclei. Such a high track density indicates that in these crystals the Th-U nuclei track density can reach 10^2-10^3 $tr.cm^{-3}$.

As was mentioned above, the high background due to Fe nuclei tracks does not allow the revealing of the total etchable track length which requires a rather long etching time and results the destruction of the crystals.

Further, because of the effects of partial annealing of fossil tracks in outer space, the direct comparison of the etchable length of these tracks and of the "fresh" tracks of accelerated ions from Ti to Kr is incorrect. Moreover, from the very beginning of olivine crystals study it has been found that these crystals have rather high volume density (10^5-10^7 cm^{-3}) of the so-called capillar inclusions-linear defects with lengths between a few tens of micrometers and 1-2 mm and with widths in the region of 0.1 μm up to 1-2 μm (before etching). The etching figures of such capillar inclusions may imitate tracks due to fast ultraheavy nuclei. Later more thorough investigations /17,18/ of these capillars shows that usually they are oriented along the main crystallographical axes [001], [100], [010], [110], but sometimes one can find such inclusions along secondary orientations - such as [102], [203], [150], etc. The maps and tables which contain more thorough information on the orientation of such systems of capillar inclusions were presented in /17,18/.

In all these investigations the cosmic-ray tracks oriented parallel to these capillar defects of olivine structure, were always excluded from further study /1,2,13-19/.

In spite of all the difficulties mentioned above, at the first stage of these investigations the preliminary data on the abundancies of cosmic ray nuclei from Sn to Pt-Pb and Th-U relative to Fe group nuclei and on the energetic spectra of all the nuclei of this group (from Fe to U) were obtained /14,16,19/.

In particular, the abundance of Th-U nuclei relative to Fe nuclei was obtained equal to $(1-2)\times10^{-7}$, and $\sim4\times10^{-2}$ /15,16,19/ relative to Pt-Pb. These values correspond reasonably well to the known Solar system abundances attributed to the time of termination of the Solar system's nucleosynthesis, about 4.6 B.Y. ago.

At the second stage of investigations, from the end of 1979 the partial controlled annealing technique of tracks in crystals (prior etching) was applied /21/ for olivines from the meteorites Marjalahti and also Eagle Station. The chosen procedure of olivine crystal annealing, at $430^{\circ}C$ during 32 h, not only eliminated completely fossil tracks due to Fe nuclei - $10^{10}-10^{11}$ $tr.cm^{-3}$, but also resulted in the shortening down of tracks due to $Z\geqslant50$ nuclei, by a factor of 6-8 and provided the erasing of differences in the thermal history of the tracks, which had been recorded during the outer space exposure of these meteorites, up to few tens and hundreds of M.Y. /1/.

The predicted dependence of the etchable track length on the atomic number Z of the nuclei for such annealing procedure has been supported by only one experimental point - the etchable track length due to accelerated ^{132}Xe - (26.5 ± 2.5) μm and on extrapolation of etchable track length for atomic numbers Z=80,92,110 basing on the Katz and Kobetich model /22/. The precision of these predictions was not better than $\pm15\%$.

For revealing the tracks due to $Z \geqslant 50$ nuclei completely situated inside the crystal volume, the annealed olivines were irradiated before etching with a well-focused beam of a Nd laser which produced a system of cylindrical canals with a diameter of 100 μm surrounded by radial microcracks up to 250-300 μm deep /21/.

In the first work performed with the aid of this technique in 1980 by Perelygin and Stetsenko /1/ the track length spectra (fig.1) exhibit track groups 120-140 and 190-220 μm long. These groups were attributed to the cosmic-ray nuclei lying in the region of Pt-Pb and Th-U respectively. The only very long (365 μm) track

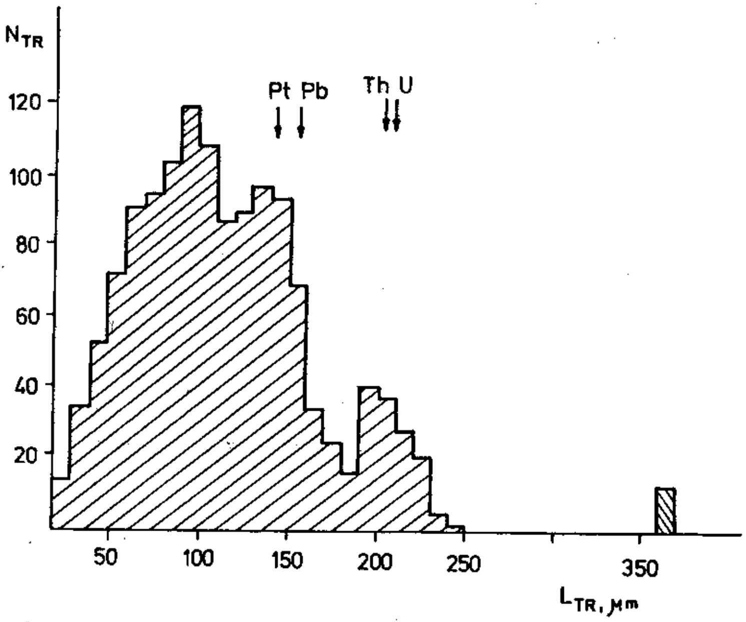


Fig. 1. The spectrum of the lengths of "fossil" tracks from galactic cosmic-ray nuclei in olivine crystals from meteorites. The crystals were annealed at 430°C during 32 hours /1/.

discovered in /1/ could have been produced by $Z \geqslant 110$ cosmic-ray nuclei, coming to rest in olivine crystals. In a further study /2/ 10 more such anomalously long tracks were measured. They form a rather compact group with a length of 340-360 μm (see fig. 2).

In paper /2/, basing on the assumption that the 340-360 μm track group was due to atomic nuclei with $Z \geqslant 110$, the abundance of

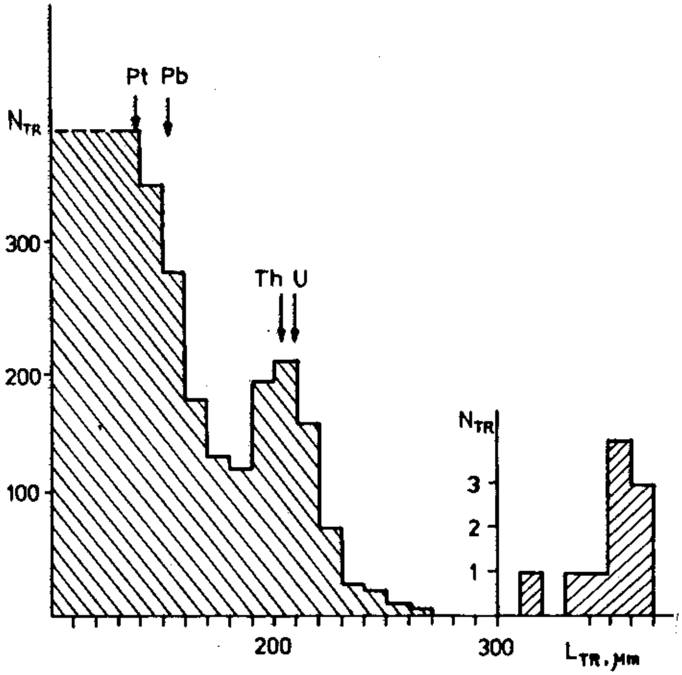


Fig. 2. The total spectrum of the lengths of "fossil" tracks from galactic cosmic-ray nuclei in olivine crystals in the period 1980-1987. The crystals were annealed at 430°C during 32 hours. 90% of the tracks were measured in crystals from the Marjalahti meteorite the rest in crystals from the Eagle Station meteorite /2/.

such nuclei relative to Th-U was estimated to be $(3\text{-}10)\times10^{-3}$. The previous search for spontaneously fissioning SHE in meteorites and in terrestrial samples /23/ gives their abundance of $<10^{-6}$ relative to U. But the direct comparison of both these values for SHE abundances is incorrect because even the average age of cosmic rays (10^7 years) is much smaller than the age of the Solar system (~ 4.6 B.Y.). The above presented value of abundance, $(3\text{-}10)\times10^{-3}$, agrees only with the most optimistic estimates of nucleosynthesis of SHE in the r-process.

From late in the 70's to the early 80's more pessimistic estimates appeared, which took into account the "cut off" in r-process nucleosynthesis due to the process of delayed fission, and predicted either the lowering of the yield of superheavy nuclei by factor 10^2-10^3 /24,25/ or even completely excluded synthesis of SHN /26/.

The disagreements in this kind of calculations and estimates show that the origin of the groups of the 120-140 μm, 190-220 μm and especially 340-360 μm cosmic-ray nuclei tracks is very important for better understanding of galactic nucleosynthesis. For the correct identification of the cosmic-ray nuclei Pt-Pb and Th-U, and for the establishment of the origin of the anomalously long tracks it was necessary to perform the calibrations of olivine crystals with accelerated heavy nuclei in the region $Z\approx 80$-92 and with energy $E > 25$ MeV/nucl.

Such heavy accelerated ions one can get only at the Bevalac accelerator facility, Lawrence Berkeley Laboratory, California.

The calibration of a few hundred olivine crystals from meteorites Marjalahti and also Eagle Station with ^{238}U nuclei was performed in joint experiment at the Bevalac accelerator in November 1987.

The olivine crystals varied in dimension from 1 to 5 mm. They were obtained from bigger single crystals extracted from a pallasite matrix by a shocking procedure. These crystals were mounted in epoxy 10 to 30 olivines in each mount with subsequent grinding and polishing. The bombardment of olivine crystals was performed with ^{238}U ions with energies 30 and 70 MeV/nucl, the angle of incidence of these ions was 25^{o} to the surface. A number of mounts were exposed twice, at 25^{o} and 10^{o} to the surface.

The ^{238}U beam was 15 cm in diameter, with a total intensity 10^{3} ion.sec^{-1}. Such dimensions of the beam provided simultaneous exposure of up to 20 mounts of olivine crystals each mount having a diameter of 25 mm. The fluence of uranium ions at the olivine crystals surface was $(1.0\pm0.2)\times10^{4}$ nucl.cm^{-2}. The flux and energy of the uranium nuclei in these exposures was measured with the aid of two scintillation counters, using the time-of-flight method /27/. This fluence was also controlled by muscovite mica detectors exposed together with olivine crystals at the same angles of incidence.

The annealing of olivines, containing latent tracks due to ^{238}U nuclei, at $430^{o}C$ during 32 hours and all other procedures of their development and scanning under a microscope were exactly the same, as in previous experiments on the study of galactic cosmic ray nuclei /1,2/.

As in those former experiments, the tracks of ^{238}U nuclei were etched due to intersection with only one microcrack formed by a focused Nd laser beam /1,2,21/.

The etched crystals were scanned under an optical microscope with magnification 640X. Only those U nuclei tracks, both the beginning and the end of which were situated under the crystal surface and inside its volume, were measured. The width profile of the etched ^{238}U tracks was quite similar to those of the fossil track group with the length ~ 210 μm.

The results of our measurements for 83 crystals of Marjalahti meteorite are presented in fig. 3. The analysis of the track length spectra due to ^{238}U (fig. 3) and the fossil track length spectra in

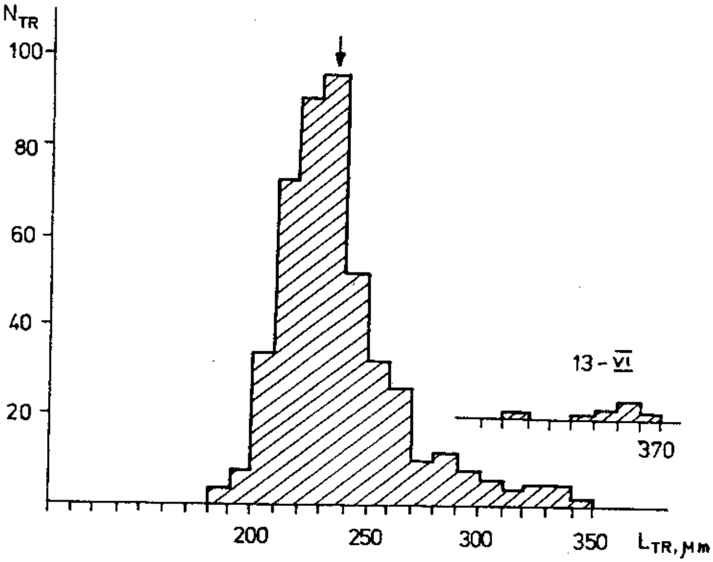


Fig. 3. The spectrum of the tracks due to accelerated ^{238}U nuclei in olivine crystals from the Marjalahti meteorite. The crystals were annealed at 430°C during 32 hours before etching. The inset shows the ^{238}U track group oriented along the [102] direction of the crystal.

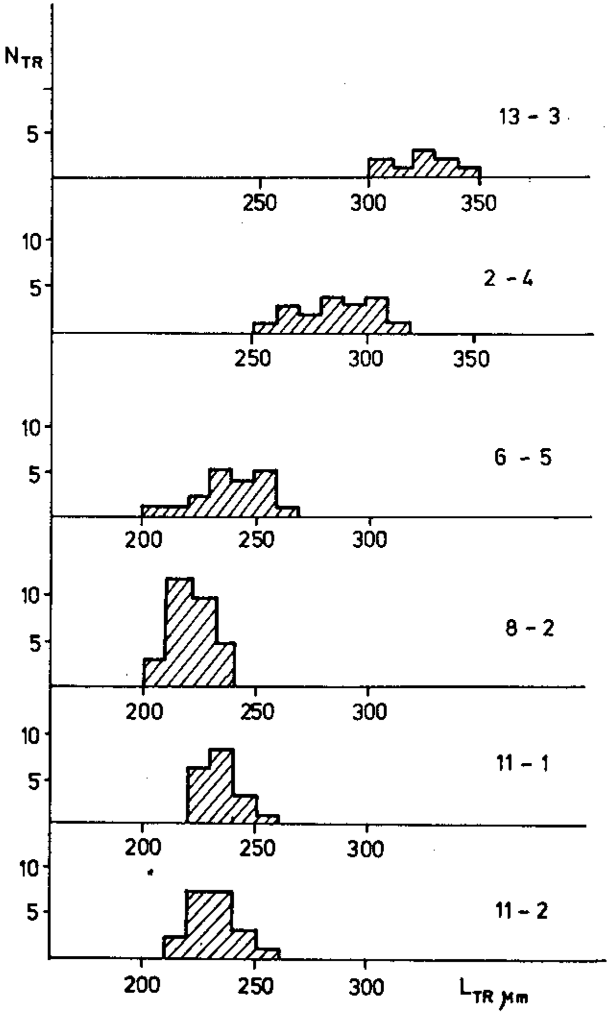
the region of (210 ± 20) μm (fig. 1,2) displays the similarity of the shapes of both these spectra.

The position of the maximum track length spectra of ^{238}U for Marjalahti crystals (230^{+25}_{-20}) μm (fig. 3) agrees with those for fossil galactic cosmic ray tracks (fig. 1,2) within the shown error bar. Some systematical shift of the mean track length due to ^{238}U to the mean fossil track length due to Th-U nuclei (fig. 2) - about 10-15% - could be explained by very long (up to 180 M.Y.) periodical heating of Marjalahti meteorite when this meteorite goes closer to the Sun. For such long time intervals the effects of nonthermal processes of recombination of the track structure can also take place, especially in the more diffuse high-energy part of fossil tracks. The most significant difference between the track length spectra due to ^{238}U (fig. 3) and those due to galactic cosmic-ray nuclei (figs. 1,2) is the presence of the "tail" of rather long tracks in the spectra shown in fig. 3.

The longer tracks of this "tail" were measured mainly in the crystals which were bombarded at 10° to the surface (fig. 4). The splitting of olivine crystals as a result of their perfect cohesion

Fig. 4. Distributions of the lengths of the ^{238}U tracks in six crystals from the Marjalahti meteorite (represented by the spectrum shown in fig. 3). Two upper histograms exemplify the presence of prolonged ^{238}U tracks which is seemingly associated with the close coincidence of orientation of these tracks with one of the major crystallographic planes in olivine.

in the crystal plane (010) led to the fact that during further operations - mounting in epoxy resin, grinding and polishing - the external polished surface was nearly parallel to the main crystal plane (010) in about 30% of the crystals. In so oriented crystals



the effect of prolongation of ^{238}U tracks has just been observed. In accordance with the data of our preliminary analysis, the ^{238}U track length distribution changes from one crystal to another depending on the orientation of the main crystallographic axes. On the other hand, for each given orientation in crystals such a length distribution seems to be rather narrow (fig. 4).

From fig. 4 one can see that the mean ^{238}U track length in some crystals exceeds by 30-40% the mean track length for most of olivines (95%) from Marjalahti meteorite. In one of the crystals ^{238}U tracks as long as 330-370 μm were measured. But in parallel direction (with precision $\pm 1^{\circ}$) in this crystal the linear etching figures are observed due to defects oriented along crystallographic direction [102]. As has been pointed out above, the heavy nuclei tracks parallel to such defects of structure were not considered in our study.

Another proposed explanation of this effect was the considerable difference in chemical composition of a part of the crystals investigated. In order to clear out the situation, we determined the chemical composition of 6 crystals in some of which the usual, and in other prolonged tracks of ^{238}U nuclei were measured. The measurements were carried out by the method of X-ray electronic microanalysis on the Link System 860-500 facility /28/. Within the measurement (±4%) error for this method the chemical composition of all these crystals was found to be the same.

For the purpose of a more detailed study of the thermal stability of ^{238}U tracks in olivines we carried out experiments on annealing such crystals during 32 h at temperatures of 450°C, 440°C, 436°C and 410°C. In those annealing procedures only crystals irradiated at 25° to the surface were used. In each group a minimum of 60 olivine crystals were annealed. The results of this annealing study are presented in figs. 5-7. For annealing at temperatures 450°C and 440°C the mean ^{238}U track length is 120^{+25}_{-15} µm and 190^{+20}_{-15} µm respectively (see figs. 5 and 6). The annealing at 436°C results in the shortening down of ^{238}U nuclei tracks to the value 210^{+25}_{-20} µm (fig. 7), which is closest to the spectra of fossil tracks due to Th-U cosmic-ray nuclei (figs. 1,2). The change in annealing temperature from 450°C to 410°C results, as is clear from fig. 8, in a smooth decrease in the mean etchable track due to ^{238}U nuclei.

Turning to the analysis of the results obtained, one can conclude that, first, the fossil tracks with a mean length 210 µm (figs. 1,2) have been formed from the galactic cosmic-ray nuclei group of Th-U.

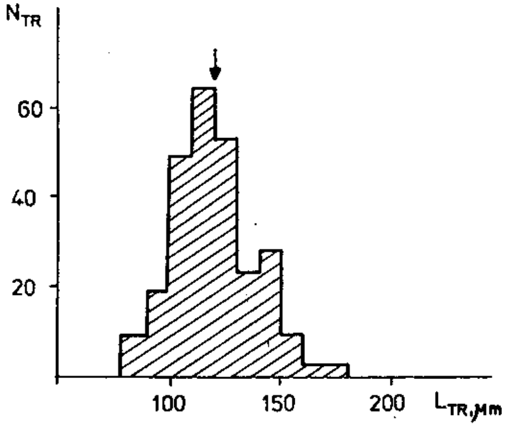


Fig. 5. Distributions of the lengths of the ^{238}U tracks in crystals from the Marjalahti meteorite. The crystals were annealed at 450°C during 32 hours.

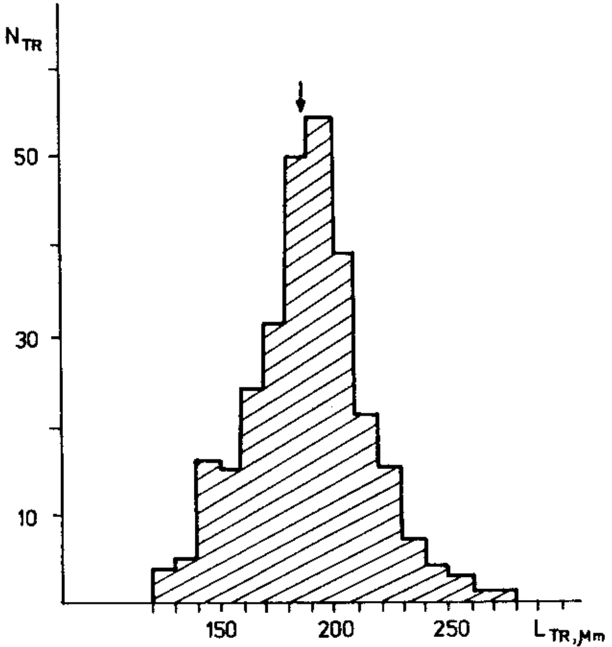


Fig. 6. Distributions of the lengths of the ^{238}U tracks in crystals from the Marjalahti meteorite. The crystals were annealed at 440°C during 32 hours.

Second, the presence in the calibration spectra due to ^{238}U (fig. 3) of tracks with a length of up to 350 μm (i.e. very close to the group of 340-360 μm fossil tracks) means, that any strict explanation of the origin of anomalously long fossil tracks is nonconclusive and needs a further study and analysis to be made. Similar rather controversial results on the etchable track length in olivines calibrated by ^{238}U ions and annealed under the same annealing conditions were published recently /29/.

For further studies of the origin of the group of the longest fossil tracks it is necessary to examine more closely the dependence of the etchable length of the accelerated Au, Pb and U nuclei tracks on their orientation in the olivine crystal lattice.

Such a study allows one to find out the possible crystallographic directions and regions of the solid angles in which the enlarging of etchable track lengths can take place.

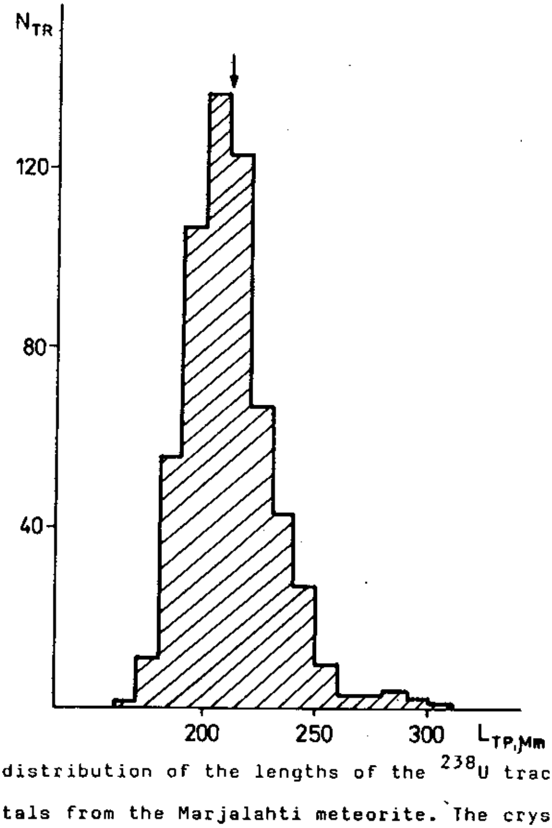


Fig. 7. The distribution of the lengths of the ^{238}U tracks in crystals from the Marjalahti meteorite. The crystals were annealed at 436°C during 32 hours before etching.

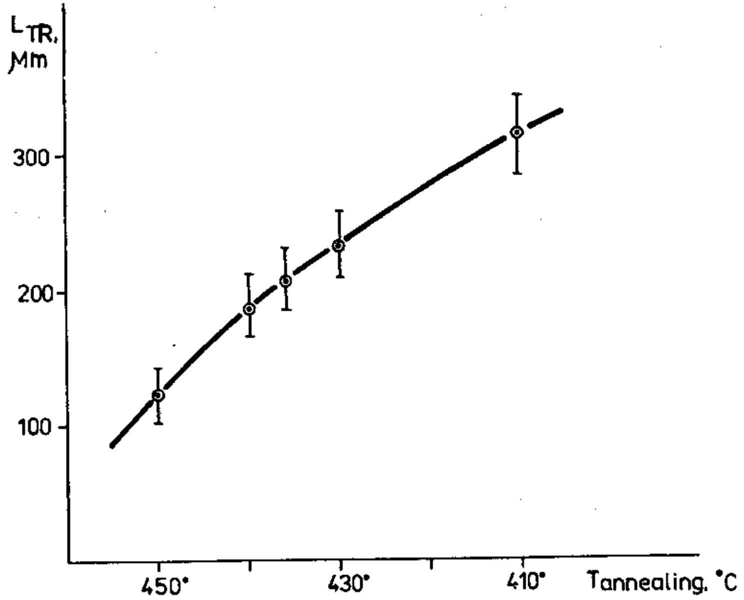


Fig. 8. Dependence of the mean etchable lengths of the ^{238}U tracks on annealing temperature in the range 410-450°C. The annealing was performed during 32 hours.

Such data can be directly compared with the known orientation of each anomalously long track. Further it can be pointed out that more detaled measurements of the lengths and profiles of both "fossil" and "fresh" tracks as functions of their orientation in olivine crystals will give more precise information about the dependence of the etchable track length on the atomic number of cosmic-ray nuclei and thus improve the resolving power of the method used.

In conclusion the authors express their deep gratitude to Academician G.N.Flerov for suggesting the method, for thoughtful

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