Pramāṇa, Vol. 20, No. 4, April 1983, pp. 287-292. © Printed in India.

A search for superheavy nuclei tracks in extraterrestrial olivine crystals

J S YADAV, A P SHARMA*, G N FLEROV**, V P PERELYGIN**, S G STETSENKO**, P PELLAS+, C PERRON+, R ANTANASIJEVICH++, B JAKUPI++ and Y TODOROVICH++
Tata Institute of Fundamental Research, Bombay 400 005, India
*Physics Department, Kurukshetra University, Kurukshetra 132 119, India
**Joint Institute for Nuclear Research, Dubna, Head P.O. Box 79, Moscow, USSR
+Laboratorie de Mineralogie du CNRS Rue Buffon 61, Paris 75005, France
++Institute of Physics, 1100 Belgrade, Yugoslavia

MS received 13 August 1982: revised 12 February 1983

Abstract. A study is made for the search of superheavy nuclei in Marjalahti, Eagle Station and in other pallasite olivines. The olivine crystals are calibrated for heavy ion track lengths by using heavy ion beams from cyclotrons. The calibration for ultra heavy ions which are presently not available with sufficient energy to produce volume tracks in olivine crystals, is based on Katz and Kobetich model of track formation. The length spectrum of volume tracks, revealed by puncturing them with focussed Nd-glass laser beam, is measured and the abundances of different nuclei groups are calculated. Partial annealing has been used at 430°C for 32 hr which eliminates the interfering tracks due to nuclei of atomic number $Z \leq 50$. During the scanning 4 cm^3 olivine crystals, about 360 long tracks of uranium group as well as two very long tracks have been found. If these tracks belong to superheavy nuclei, the relative abundance of super heavies is found to be 6×10^{-11} in galactic cosmic rays.

Keywords. Meteorites; volume tracks; galactic cosmic rays; superheavy nuclei.

1. Introduction

Experiments to search superheavy nuclei were started by Fowler *et al* (1970) in stacks of thick layers of nuclear emulsions exposed to galactic cosmic rays by using balloon flights. The main problem with such experiments (using direct registration of ultra-heavy nuclei from galactic cosmic rays) was a very low flux of $Z \geqslant 30$ nuclei in the cosmic rays. Therefore, in order to increase the statistics in such experiments it is necessary to expose large areas of these detectors in space for a long time. In experiments with nuclear emulsion and high polymer dielectric track detectors, only 23 tracks due to actinides have so far been observed and not a single track was found which could be attributed to nuclei of $Z \geqslant 110$ (Fowler *et al* 1977; Shirk and Price 1978). The detailed analysis of these results shows that the identity of tracks due to nuclei of atomic number lying between 76 and 100 does not possess good statistics and moreover there may be a possibility of overestimating the atomic number of cosmic ray nuclei in this region (Meyer 1979).

Another possibility of search for superheavy nuclei in galactic cosmic rays is connected with the silicate crystals from meteorites which have been exposed for a period of several million years in space (Maurette *et al* 1964). The advantages of using silicate crystal detectors from meteorites are that (i) they register nuclei with $Z \geqslant 20$ only, (ii) they are less prone to annealing which may occur during their space life and (iii) they are exposed in free space for millions of years. The density of tracks due to Th-U nuclei is estimated to be up to 10^4 tracks/cm^3 and up to 10^3 tracks/cm^3 in the silicate crystals located at a depth of $\leqslant 1$ cm and $\leqslant 5$ cm respectively, from the pre-atmospheric surface of the meteorites exposed for more than 10^8 years. The silicate crystals from pallasites are most suitable for the study of VVH cosmic ray abundance as (a) crystals of big size are available in pallasites with good transparency, (b) the chemical composition of pallasitic olivine is homogeneous within the same pallasite and, (c) the exposure ages of pallasites are in general higher than those of stony meteorites. For the above reasons we have selected Marjalahti, Eagle Station and other pallasites for our study. Moreover the olivines are embedded in a nickel—iron (Ni-Fe) matrix in pallasites; hence the space erosion of these meteorites should be less as compared to lunar rocks and stony meteorites.

In the first stage of our investigation, about 20 stony-iron pallasites were surveyed for high VH track density. From these pallasites Marjalahti, Lipovsky Khutor and Eagle Station (possessing locations 2 to 6 cm below the preatmospheric surface) were selected for further investigations of superheavy nuclei (Otgonsuren *et al* 1976). The olivine crystals were used for calibration of heavy ion track lengths (from Ti to Xe) by using accelerated heavy ion beams from cyclotrons (Perelygin *et al* 1977). The etching figures of capillar inclusions and dislocations in olivine crystals were studied extensively so as to distinguish them from nuclear particle tracks (Dolivo-Dobrovolskaya *et al* 1976). In earlier experiments of Lhagvasuren *et al* (1980a) more than 5000 tracks due to $Z \geqslant 50$ cosmic ray nuclei were measured in 0.7 cm^3 meteoritic olivine. About 150 tracks longer than 700 μm were attributed to Th-U group.

In the present study about 4 cm^3 olivine crystals from Marjalahti and Eagle Station pallasites are annealed at 430°C for 32 hr. The length spectrum of volume tracks is measured. The abundances of different nuclei groups are calculated from volume track length spectrum using our L-Z identification method.

2. Experimental procedure

To accelerate the search for superheavy nuclei tracks we have used the method of partial annealing of tracks (Kapuscik *et al* 1966). This method corresponds to an increase in the threshold for revealing the tracks and hence decreases the length of tracks of VVH nuclei and erases completely the tracks due to low ionizing particles. Thus partial annealing permits use of abundant crystals of small size and possibly extends the measurements to other meteorites in which large crystals are not found. The annealing behaviour of tracks of different heavy ions Cr, Fe, Ge, Kr, Xe, Pb and U are studied in detail and suitable annealing conditions are inferred. At temperatures $\geqslant$ 480°C the annealing behaviour of Xe, Pb and U tracks is found to be irregular (Lhagvasuren *et al* 1980b). The annealing at 430°C for 32 hr erases the tracks completely due to nuclei of $Z \leqslant 50$ and hence reduces the surface track density (Yadav 1982). This allows us to handle crystals located near the surface of meteorites.

The olivine crystals are etched in WO_4 solution. For revealing volume tracks in olivine crystals we have used the focussed Nd-glass laser beam which produces holes accompanied by systems of cracks. It may be mentioned here that this procedure provides more effective revealing of tracks with $l \geqslant 150\ \mu m$ than of tracks with $l < 150\ \mu m$ (Yadav 1982).

3. Results and discussion

The optimal annealing conditions for partial annealing has been obtained from our detailed annealing study and is found to be at 430°C for 32 hr. All the tracks due to nuclei $Z \leqslant 50$ are eliminated completely under these annealing conditions. The tracks length of Xe ions from accelerator has been measured and found to be $(26 \pm 3{\cdot}5)\ \mu m$ for these annealing conditions. The total etchable track length for very heavy nuclei which are not available at present with sufficient energy to produce volume tracks in crystals, has been calculated theoretically as a function of atomic number and is shown in figure 1 (Katz and Kobetich 1968). The experimental points for accelerated Fe, Zn, Ge and Kr ions are also shown. In figuie 1, the etchable length of tracks after annealing at 430°C for 32 hr has also been shown as a function of atomic number. The etchable track lengths are estimated for various groups of nuclei. It is found to be 140-180 μm for Pt-Pb group, 180-240 μm for Th-U group and 350-400 μm for nuclei $Z \sim 110$. This criterion has been used for the assignment of charge to etched tracks from galactic cosmic rays in olivine crystals.

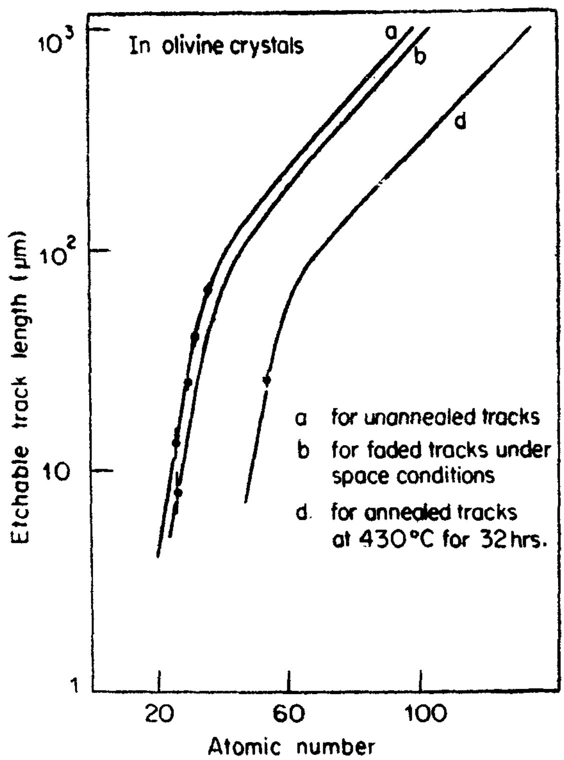

Figure 1. Dependence of etchable track length on atomic number in olivine crystals.

The results of our measurements of the volume track lengths in about 3 cm^3 olivine from Marjalahti are presented in figure 2. Three hundred long tracks of length $180 \leqslant l \leqslant 240$ μm are attributed to actinide nuclei on the basis of our L-Z semi-empirical estimation. In addition, a very long track of length 365 μm is also found during our measurements. The track length spectrum of volume tracks measured in 1 cm^3 olivine crystals from Eagle Station is presented in figure 3. About 60 long tracks which can be attributed to the uranium group and a very long track of 350 μm have been found. The track length of two long tracks is higher than the mean etchable track length of Th-U unclei by a factor of 1.5–2, hence these tracks cannot be included in this gıoup. Thus these tracks may be attributed to nuclei with Z $\sim$ 110.

The length of the conical part of tracks of various heavy ions has also been measured and found to be $(11{\cdot}5 \pm 2)$ μm for Xe ions and $(67{\cdot}5 \pm 5)$ μm for U group tracks. In the case of these two long tracks the length of conical parts is (120 ± 10) μm and (110 ± 10) μm respectively, which agrees with the supposition that these tracks are due to superheavy nuclei ($Z \sim 110$) (Yadav 1982). We have also studied

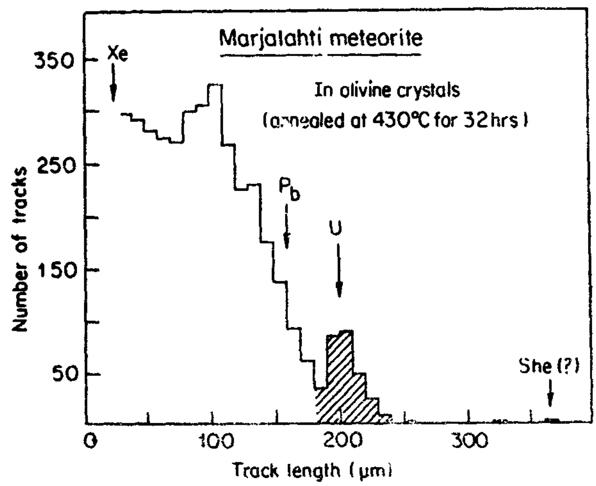

Figure 2. Track length distribution in Marjalahti olivine crystals for annealing at 430°C for 32 hr.

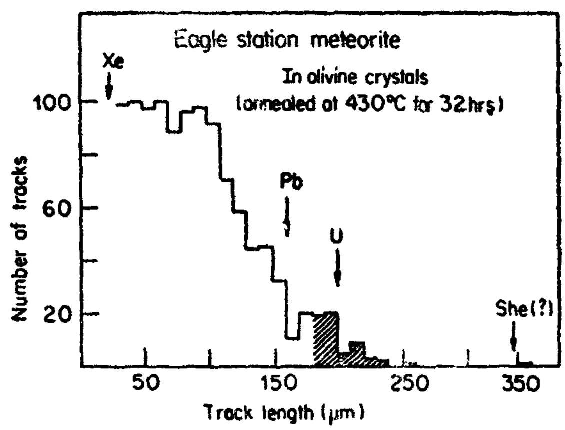

Figure 3. Track length distribution in Eagle Station olivine crystals for annealing at 430°C for 32 hr.

Table 1. Abundances of VVH cosmic rays relative to Fe-group.

Charge groups	Abundance calculated from our measurements (meteoritic crystals)	Solar system abundances
$57 \leqslant Z \leqslant 62$	2.8×10^{-6}	3.2×10^{-6}
$62 \leqslant Z \leqslant 74$	8.7×10^{-6}	2.0×10^{-6}
$74 \leqslant Z \leqslant 83$	4.6×10^{-6}	8.7×10^{-6}
$90 \leqslant Z \leqslant 96$	9.7×10^{-7}	1.0×10^{-7}

the effect of partial annealing on the length of the conical part of Th-U nuclei. About 1 g olivine is annealed at (382 ± 2)°C for 72 hr. For these annealing conditions the Kr ion tracks are shortened down to (18 ± 3) μm, but our measurements show that the length of the conical part for U-group tracks (which have a total track length of 380-430 μm) after annealing remains the same.

To ensure that these two tracks are due to nuclear charged particles the orientation of these tracks has been measured with respect to the main crystallographic axes of olivine crystals. We have found that the orientation of tracks does not coincide with primary or secondary crystallographic directions. This excludes the possibilities of both the channeling effect which increases the etchable track length and of etching figures due to capillar inclusions in olivine crystals (Dolivo-Dobrovolskaya *et al* 1976).

We have scanned 4 cm^3 olivine crystals from meteorites. Abundances of different charge groups relative to Fe-group are presented in table 1. The solar system abundances are also shown in table 1 for comparison (Cameron 1973). The results show general agreement except for the uranium group nuclei which shows a drastic increase by a factor of ~ 10. Recent experiments with lexan detectors have also shown that uranium group nuclei are over-abundant by a factor of ~ 9 in galactic cosmic rays compared to solar system (Shirk and Price 1978). These results show that cosmic ray sources are greatly enriched in uranium group nuclei if our charge assignment is correct.

We have also studied olivine crystals from Luna-16 and Luna-24 probes. The track density of the Fe group nuclei is found up to $\geqslant 10^8$ tracks/cm^2 in these crystals. In some of these crystals the track density due to $Z \geqslant 30$ nuclei is found to be up to 10^5 tracks/cm^2 which is higher than that in meteoritic olivine (Yadav 1982). Further investigations with lunar olivine crystals are in progress.

References

Cameron A G W 1973 *Space Sci. Rev.* **15** 201

Dolivo-Dobrovolskaya G I, Kalamensky V D, Gavrilova N N, Perelygin V P and Stetsenko S G 1976 *Geokhimiya* **10** 1476

Fowler P H, Adams R A, Cowen V G and Kidd J M 1970 *Proc. R. Soc.* **301** 35

Fowler P H, Alexandre C, and Clapham V M 1977 *Proc. 15th Int. Cosmic Ray Conf. Plovdiv* **11** 165

Kapuscik A, Perelygin V P, Tretiakova S P and Shadieva N H 1966 *Proc. 6th Int. Conf. on Corpuscular Phot.*, Florence CEP, Roma 458

Katz R and Kobetich E I 1968 *Phys. Rev.* **170** 402

Lhagvasuren D, Otgonsuren O, Perelygin V P, Stetsenko S G, Pellas P and Perron C 1980a Reprint from Solid State Nuclear Track Detectors 997

Lhagvasuren D, Perelygin V P, Stetsenko S G and Murtazaev K H 1980b Reprint from SSNTD 929

Maurette M, Pallas P and Walker R M 1964 *Nature* (London), **204** 821

Meyer J P 1979 *Proc. 16th Int. Cosmic Ray Conf. Kyoto* **1** 374

Otgonsuren O, Perelygin V P, Stetsenko S G, Gavrilova N N, Fieni C and Pellas P 1976 *Astrophys. J.* **210** 258

Perelygin V P, Stetsenko S G, Pellas P, Lhagvasuren D, Otgonsuren O and Jakupi B 1977 *Nucl. Track Detection* **1** 199

Shirk E K and Price P B 1978 *Astrophys. J.* **220** 719

Yadav J S 1982 *Study of charged particle tracks in* SSNTDs Ph.D. thesis, Kurukshetra University, India.