A Long Term Change in the Cosmic Ray Composition?: Studies on Fossil Cosmic Ray Tracks in Lunar Samples

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Phil. Trans. R. Soc. Lond. A 1977 **285**, 593-599
doi: 10.1098/rsta.1977.0104

Phil. Trans. R. Soc. Lond. A. **285**, 593–599 (1977)

Printed in Great Britain

A long term change in the cosmic ray composition?: studies on fossil cosmic ray tracks in lunar samples

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The etching techniques for the identification of very heavy cosmic ray ions from their etchable tracks in mineral track detectors are described and the results so far obtained for the ancient galactic cosmic ray Cr group (V + Cr + Mn) to Fe abundance ratio are presented. It was found that the etchable radiation damage of fossil cosmic ray tracks has probably only been slightly affected by annealing processes. The track data obtained on pyroxenes of different lunar rocks and on pyroxenes and feldspars, i.e. detectors of different track retaining characteristics, yielded consistent results. From this measurements, an ancient Cr group to Fe ratio of approximately 0.7–0.8 was deduced. In comparison with the present day galactic cosmic ray composition, this ratio is enhanced by a factor of about two. From the track data obtained in different lunar soil samples it was concluded that a variation in the Cr group to Fe ratio between 0.4–0.8 exists. Both results indicate, that either a long term change in the cosmic ray composition has taken place or the interpretation of track data is much more complicated than assumed.

INTRODUCTION

Certain mineral crystals (e.g. pyroxenes and feldspars) are able to store the etchable radiation damages of very heavy ions for geological time spans. After suitable etching, the damaged structure of the crystal is preferentially dissolved and a track – i.e. an etched channel along the ion path – is generated. These tracks can be easily studied by means of an optical microscope.

Lunar and meteoritic samples have been subjected to cosmic ray bombardment for millions of years. Thus track detecting minerals of these samples provide the unique opportunity to gain information for example about the long term history of the abundances of very heavy ($Z \geqslant 20$) ions in the cosmic radiation (see, for example, Price & Fleischer 1971; Price *et al.* 1973). The results thereby obtained necessarily represent averages over the exposure times of the samples. For lunar samples, these times are of the order of several million years (Crozaz *et al.* 1974). First charge assignments to cosmic ray tracks in pyroxenes of the Patwar meteorite were given by Lal (1969). Similar investigations were then performed on olivines and pyroxenes of various lunar and meteoritic samples (Lal *et al.* 1970; Bhandari *et al.* 1971; Bhandari & Padia 1974). By a direct comparison of cosmic ray tracks and tracks of artificially accelerated heavy ions in lunar pyroxenes an attempt was made to base the charge assignments on experimental results (Plieninger *et al.* 1972, 1973). Furthermore, ion identification methods for feldspars were developed and cosmic ray tracks were studied in lunar feldspars (Krätschmer & Gentner 1975).

The main results of these investigations are the following: The ancient cosmic ray $Z = 30$–40 to $Z = 21$–28 abundance ratio seems to have been approximately constant over the last few billion years (Bhandari & Padia 1974). Concerning the $Z = 20$–28 range, in many, but not in all charge distributions so far obtained for lunar samples no such prominent Fe peak has been observed as one would expect from the abundances of these ions in the present day cosmic radiation. This has been regarded as an indication for a long term variation in the cosmic ray composition, especially in the Cr/Fe abundance ratio (Plieninger *et al.* 1973).

In this work, a survey of ion identification techniques will be given and the present status of investigation into the Cr/Fe abundance ratio of the ancient cosmic radiation will be presented.

ION IDENTIFICATION TECHNIQUES

The radiation damage generated by a heavy ion along its path through the crystal results in an enhanced etching rate along the ion trail. Both, radiation damage and track etching rates, increase with decreasing residual range and reach a maximum near the stopping point of the ion. For each ion a characteristic relation between track etching rate and residual range exists in such a manner that for a given residual range the track etching rate increases with the atomic number of the track-forming ion. Thus, by measuring both quantities, ion identifications can be made.

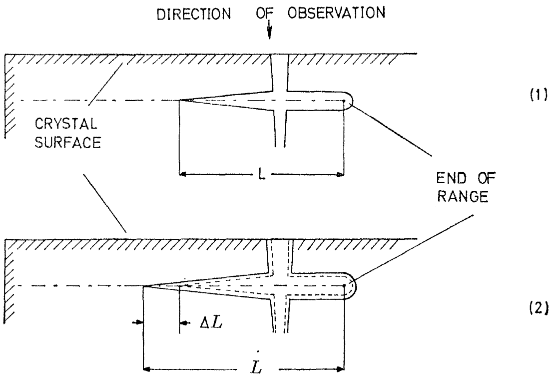

FIGURE 1. The principle of the ion identification techniques is shown on this schematical sketch, depicting an etched track in a track detecting mineral crystal after subsequent etching processes (1) and (2). As indicated, surface crossing tracks are serving as channels for the etchant. For ion identification, the track etching rate has to be determined as a function of residual range. The track etching rate can be deduced from the increase of track length ΔL. The total track length L finally reached can be regarded as a measure of the corresponding residual range.

The principle of the identification technique is depicted in figure 1. The track etching rate can be determined from the increase in track length between subsequent etching processes. The total track length finally reached after the second etching process can be regarded as a measure of the corresponding residual range. As indicated in this figure, the tracks are etched via etching channels which are provided by tracks reaching the crystal surface. Tracks developed by this procedure will be referred to as TINT (track in track) (Lal 1969). Etching channels can be generated artificially by irradiating the crystals with very heavy ions (e.g. 10 MeV/n Kr). Generally the etching times for TINT are not constant but statistically distributed according to the probability of intersection between tracks and etching channels. Thus a certain scatter results in the widths and lengths of TINT because these quantities are functions of etching time.

It was found experimentally that in comparison with feldspars the track etching rate in pyroxenes decreases very rapidly with residual range (see, for example, Krätschmer *et al.* 1973). Therefore, in this mineral the length of a track etched for a sufficiently long time already defines the atomic number of the track-forming ion. To decide whether a track has been etched for a sufficiently long time, the track width can be used as a criterion; this however is sometimes ambiguous. The track width is not only a function of etching time but also depends on the orientation of the track with respect to the crystallographic directions since the etching rates for the bulk material reflect the anisotropy of the crystal. These difficulties can be overcome by a direct comparison of the lengths of cosmic ray tracks and those tracks produced by artificially accelerated calibration ions (e.g. 10 MeV/n Fe) in the same crystal. Under these conditions, the etching times for the TINT of both kinds of ions are equal on the average. Thus for pyroxenes, the position and width of the length distribution of calibration tracks can be regarded as an indicator for the position and width of the corresponding cosmic ray ion tracks.

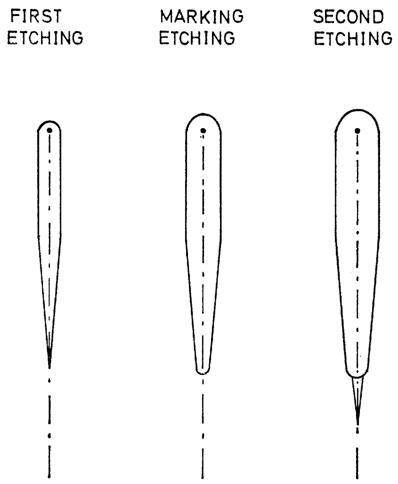

FIGURE 2. The shape of a track, resulting after each etching process, when the marking method is applied. By marking the track, the determination of the track etching rate can be simplified.

Because of the much more gradual decrease of track etching rates with residual range in feldspars, the track lengths in this mineral do not reveal the charges of the track-forming ions with sufficient accuracy. Thus, a measurement of both, track etching rate and residual range, is necessary for a more accurate ion identification. To simplify the track etching rate measurement, a marking method has been developed (Krätschmer *et al.* 1973). For this purpose the etching procedure has to be slightly modified. After a first regular etching, the high energy end of the track is marked by means of a special etchant, which reacts much less vigorously with the radiation damage along the ion path. Thereby the track becomes broader according to the bulk material etching rates of the crystal, but not significantly longer. The resulting track shapes are sketched schematically in figure 2. A further second regular etching then reveals the track etching rate from the tail-shaped continuation of the track.

In most practical situations, the individual feldspar or pyroxene crystals of the samples are too small to achieve reasonable statistics; therefore a larger number of crystals have to be investigated and the data obtained have to be appropriately superposed. Thereby the tracks of calibration ions can be used as an indicator for the degree of influence of differences in the chemical compositions and crystallographic orientations between the individual crystals.

The etchable cosmic ray tracks have been stored in the lunar crystals for millions of years and thus are in reality 'fossil' tracks. During this time, various effects (e.g. thermal annealing) may have given rise to a partial relaxation of the etchable radiation damage. Therefore one should expect that the relatively fresh calibration tracks and the fossil tracks are not directly comparable and difficulties in proper charge assignments should result. Fortunately, the fossil tracks in lunar samples investigated so far seem to be only weakly affected by annealing processes, as will be shown later on.

Experimental procedures

Pyroxene and feldspar containing lunar samples have been mounted in epoxy and polished. For calibration, the mounted sections have been irradiated at the LINAC of the University of Manchester with 10 MeV/n Fe ions at grazing incidence (about 10–20° to the sample surface). To generate effective etching channels for TINT formation, an additional perpendicular exposure to 10 MeV/n Kr ions was performed. Feldspars and pyroxenes were etched by a boiling aqueous NaOH solution with concentrations of 7 and 50 % by mass respectively. The etching times ranged from about 15–20 h for feldspars to 5 h for pyroxenes. For marking the tracks in feldspars, a 0.5 % HF solution at room temperature was used and etching times of 10–15 min were applied. After etching, the optical contrast of the tracks was enhanced by silver decoration (MacDougall *et al.* 1971). The tracks were scanned by an optical microscope.

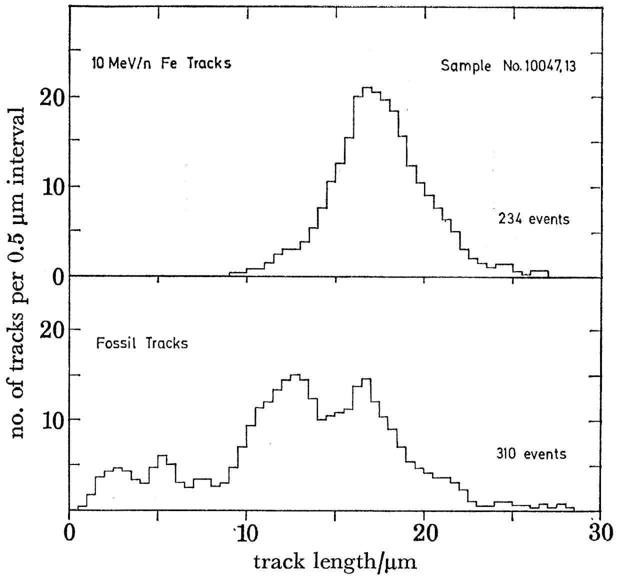

FIGURE 3. Length distributions of artificially generated Fe and fossil galactic cosmic ray tracks measured simultaneously in the same crystals of rock sample 10047. From the shape and position of the distribution of Fe calibration tracks, the ancient cosmic ray Cr group (V+Cr+Mn) to Fe abundance ratio can be estimated to about 0.7–0.8. This ratio is significantly higher than the generally accepted present day value.

Results and discussion

By direct comparison of fossil cosmic ray and Fe calibration tracks in pyroxenes of lunar rock sample 10047 it was observed that the length distribution of calibration tracks coincides with a peak in the length distribution of cosmic ray tracks at about 17 μm, as it is shown in figure 3. Preliminary data obtained by a corresponding measurement on pyroxenes of rock samples 15076 yielded a quite similar pattern: a peak at about 17 μm which approximately coincides with the peak in the length distribution of Fe calibration tracks, and a relatively large number of fossil tracks in the 10–15 μm length range.

All pyroxene crystals from lunar rock and soil samples so far investigated show a prominent peak in the length distribution of fossil tracks at about 16–18 μm, despite the fact that these different samples certainly have rather different exposure histories. These results and the observation that the 17 μm peak coincides with the peak in the length distribution of Fe calibration tracks, in our opinion suggest that (*a*) the 17 μm peak is due to cosmic ray Fe ions and (*b*) annealing effects on fossil tracks in these pyroxenes studied are of minor importance. Therefore, the large number of fossil tracks in the 10–15 μm region are very probably not partially annealed cosmic Fe ion tracks. In a previous study it was shown that under similar etching conditions Ca tracks in lunar pyroxenes yield a length of approximately 5 μm (Plieninger *et al.* 1972). Therefore, the 10–15 μm long fossil tracks should be ascribed to cosmic ions belonging to the Cr group (i.e. V + Cr + Mn). From the shape of the distribution of Fe calibration ions, the ancient cosmic ray Cr group to Fe abundance ratio can be estimated to be about 0.7 for lunar rock sample 15076 and about 0.8 for sample 10047. As can be deduced from the shielding of the crystals studied, the abundance ratios refer to an energy range of a few 100 MeV/n and thus are mainly due to the galactic component. The deduced ancient Cr group to Fe ratio is significantly higher than the generally accepted present day ratio of about 0.2–0.4 for energies between 50 and 800 MeV/n (see, for example, Cartwright *et al.* 1973).

Track length distributions measured in pyroxenes of lunar soil samples have shown that, depending on the sample chosen, variations occur in the relative abundance of tracks belonging to the 17 μm peak and to the 10–15 μm length domain. In terms of the ancient cosmic ray Cr group to Fe ratio this would correspond to ratios between 0.4–0.8. However, soil samples generally have a complex exposure history and therefore these track data may be regarded as less conclusive. Unfortunately, so far no lunar rock sample has been investigated from which a Cr group to Fe ratio of less than 0.7 could be deduced. The observed variation of this ratio in soil samples covers the gap between the present day and the higher ancient Cr group to Fe ratio observed in lunar rock samples, and therefore supports the assumption that a long term change in the cosmic ray composition has taken place. Conversely one can argue that these results indicate a systematic difference in track data obtained from rock samples as compared with those obtained from soil samples. This would imply that, for reasons not yet understood, the track data from pyroxenes are either not reliable or have been misinterpreted. To check this possibility the fossil cosmic ray and Fe calibration tracks in feldspars of lunar rock sample 15076 have been investigated by the marking method. As already mentioned, the track data obtained on pyroxenes from this rock indicate an enhanced Cr group to Fe ratio. Although the present day knowledge about the track stabilities in different detectors is rather limited, the data available show that feldspar and pyroxene detectors have different track retaining characteristics (for thermal stability see, for example, Crozaz *et al.* 1970). Therefore, a comparison of the track data obtained in both kinds of minerals should yield additional information about the degree of influence of annealing effects. The resulting distributions of track length increases (track etching rates) versus total track lengths (residual ranges) for fossil cosmic ray and Fe calibration ions are shown in figure 4. The distribution of cosmic ray tracks is both, broader and systematically displaced, in comparison with the distribution of calibration tracks. The displacement indicates that the fossil tracks in feldspars are partially annealed. However, disregarding this shift between the two distributions, the structure of the fossil cosmic ray track distribution is quite similar to that obtained with pyroxenes of the same rock sample, and rock sample 10047 (see figure 3): the majority of fossil tracks are close to the tracks of Fe calibration ions while a quite large number of fossil tracks are clustered in a region which probably corresponds to cosmic ray Cr group ions. Assuming equal distribution widths for fossil and calibration Fe tracks, an estimate of the Cr group to Fe ratio from the tracks in these feldspars yields a value of about 0.7, which is in agreement with the corresponding ratio deduced with pyroxene detectors from this rock sample. This shows that consistent results can be obtained for the pyroxene and feldspar detectors in the same rock sample under the assumption that annealing effects have given rise to a general shift of the distribution of fossil cosmic Cr group and Fe tracks. This shift is, according to the different stabilities of tracks in these different minerals more pronounced in feldspars as compared to pyroxenes. From the consistency of results we conclude that the track data obtained on both, pyroxenes and feldspars, are reliable.

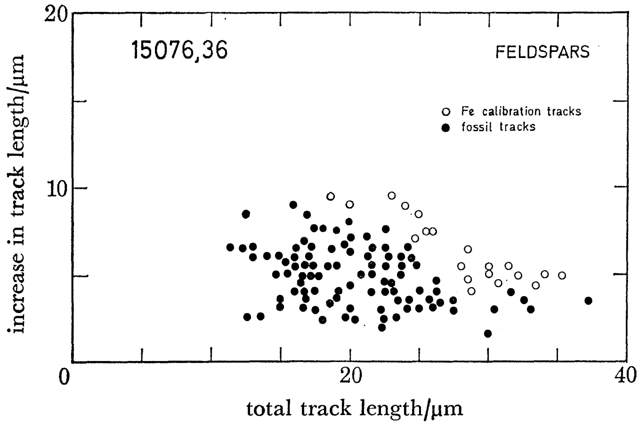

Figure 4. The distribution of track etching rates (increases of track lengths) against residual ranges (total track lengths) measured on fossil cosmic ray and Fe calibration tracks in feldspars of rock sample 15076. The fossil tracks are shortened to a certain extent. This indicates that these tracks have been affected by annealing processes. Nevertheless, the general structure of the distribution is remarkably similar to those obtained on cosmic ray tracks in pyroxenes of the same rock and on pyroxenes of rock 10047 (see figure 3).

CONCLUSIONS

The Cr group (V + Cr + Mn) to Fe abundance ratio of the ancient cosmic radiation in the energy range of a few 100 MeV/n deduced from track studies on lunar samples is approximately 0.7–0.8. This result is based on a comparison of track data obtained on (*a*) pyroxene track detectors of different lunar rock samples with certainly different exposure histories and (*b*) feldspar detectors which have in comparison to pyroxenes different track retaining characteristics to annealing processes. The consistency of the data we regard as an indication for the validity of the derived ancient cosmic ray abundance ratio. In comparison to the present day value this ratio is enhanced by a factor of about two.

The track data obtained on pyroxenes of different lunar soil samples can be interpreted with the assumption that a variation exists in the ancient Cr group to Fe ratio between 0.4 and 0.8.

The discrepancy between the present day and ancient Cr group to Fe ratio and the existence of a variation of the ancient ratio indicates that a long term change in the composition of the cosmic radiation has taken place.

There are, however, additional effects which may account for the observed enhanced Cr group to Fe ratio, e.g. nuclear fragmentation of cosmic ray Fe nuclei by interaction with the

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nuclei of the lunar matter. This would imply that the exposure histories of the lunar samples are much more complex and their average shielding depths are much larger than assumed. Further studies are necessary to solve this problem.

We are grateful to N.A.S.A. for providing the lunar samples. We thank the staff of the LINAC Laboratory (University of Manchester) for their generous support.

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