THERMAL ANNEALING OF NUCLEAR PARTICLE TRACKS IN MINERALS AND THE CHEMICAL COMPOSITION OF VERY HEAVY COSMIC RAYS. E. Dartye, J.C. Druart, J.P. Duraud, Y. Langevin, M. Maurette. Laboratoire de Physique du Solide, Faculté des Sciences d'Orsay, 91405-Orsay; Laboratoire René Bernas, 91406 Orsay; Service de Chimie-Physique, CEA, 91190 Gif-sur-Yvette.

INTRODUCTION. The determination of the relative abundance of very heavy nuclei \((Z > 30)\) with respect to that of iron-group nuclei \((20 < Z < 30)\) in the ancient cosmic rays (VWH/VH ratio) can help in solving major problems such as the origin of galactic cosmic rays and the existence of superheavy elements in nature. So far, VWH/VH ratios have been tentatively inferred from track length measurements in meteoritic and lunar material. For this purpose, the etched track length distribution, \(dN/dL\), is determined, and the track lengths are converted into various groups of atomic nuclei, by using the scaling function, \(L(Z)\), proposed by Fleischer et al. (1). However natural annealing processes have drastically reduced the lengths of fossil tracks in most extraterrestrial minerals. Thus, in order to obtain meaningful VWH/VH ratios, the scaling function \(L^*(Z)\) relevant to the partially annealed tracks has to be determined. For this purpose, we first investigated the microscopic structure of latent tracks produced by artificially accelerated ions in various minerals, as a function of both the annealing conditions and the characteristics of the incident ions, by using a small angle X-Rays scattering apparatus (SAX). The latent tracks were then etched, and their length distribution was measured with an optical microscope and a SEM. From these combined studies of latent and etched tracks, a "two-steps" annealing model was developed, which can be used to compute theoretical track length distributions that well agree with the experimental ones. Consequently, we feel confident in presenting a new method, based on our track annealing model, which is currently applied to determine meaningful VWH/VH ratios in the ancient cosmic rays.

II. EXPERIMENTAL RESULTS. Thin slabs of muscovite mica, olivine and labradorite were irradiated with Ne, Ar, Fe, Kr and Xe ions of energies ranging from 0.1 up to 10 MeV/amu. For a given mineral, two very different integrated ion doses were used \(10^{12} \text{ ions.cm}^{-2}\) and \(10^{11} \text{ ions.cm}^{-2}\). Both types of irradiated targets were submitted to isochronal and isothermal annealing runs conducted under controlled vacuum. The high dose targets were observed with the SAX whereas the low dose targets were chemically etched for the measurements of track length distributions. Our main results are: 1. SAX analysis of muscovite and olivine targets: a) latent etchable tracks are constituted of extended defects (ExD) that are linked by a chain of point defects (PtD), and the linear density \(p(\text{PtD})\) of the PtD considerably exceeds the value, \(P(\text{ExD})\), corresponding to the ExD; b) the mean size of the ExD, \(\langle \text{ExD} \rangle\), which does not sensibly depend on the ion energy and the nature of the target, increases with \(Z\). In particular in muscovite Ne and Kr ions generate \(\langle \text{ExD} \rangle\) values of 20 Å and 40 Å, respectively; c) the PtD have been observed for all ions so far investigated but the ExD are only produced by ions heavier than Ne. In addition the energy dependence of \(p(\text{PtD})\) is roughly similar to the rate of primary ionization loss, \(dJ/dx\), whereas \(P(\text{ExD})\) decreases much faster than \(dJ/dx\) in the high energy range \((E > 2 \text{ MeV/amu})\). d) Upon a thermal annealing the PtD disappear at a relatively
low temperature, \( T_c \), whereas the number of \( \Delta x \Delta D \) starts to decrease at a much higher temperature \( T_p \), in following an Arrhenius type kinetic. During annealing, the \( \langle \Delta x \Delta D \rangle \) values do not significantly change. 2. Etched track length distributions: a) Muscovite targets irradiated with Fe-ions of various energies (1.7, 3.7 and 7 MeV/amu) and fission fragments. For all ions a very narrow peak centered on the expected ion range is observed before annealing. Upon a low temperature annealing \( (T < 300^\circ C) \) the distributions due to the "low" energy ions (1.7 and 3.7 MeV/amu Fe-ions and fission fragments) are almost unchanged whereas that corresponding to the "high" energy (7 MeV/amu) Fe-ions shows an additional 15 \( \mu \)m-peak. For higher annealing temperature a transfer of tracks from the 54 \( \mu \)m-peak to the 15 \( \mu \)m-peak occurs for the 7 MeV/amu Fe-ions, and a slight broadening of all distributions in favor of short tracks is observed. We also note that the annealing temperature clearly depends on the nature of the incident ions, as fission fragment tracks start to shorten at much higher temperature \( (\sim 500^\circ C) \) than Fe-tracks \( (\sim 400^\circ C) \); b) Labradorite targets irradiated with 10 MeV/amu Fe-ions and etched with the "TINT" etching technique of Lal et al. (2) in order to reveal the so-called "maximum" etchable length of the tracks. Before annealing the maximum track length distribution appears as a single peak centered around \( \sim 56 \mu \)m, and characterized by an half-width, \( \Gamma_F \sim 10 \mu \)m. Upon a low temperature annealing \( (T < 300^\circ C) \) this distribution clearly shifts to shorter lengths \( (\sim 44 \mu \)m) while still keeping the same \( \Gamma_F \) value. At 450\(^\circ\)C, the track lengths are further reduced \( (\sim 36 \mu \)m\) but now the \( \Gamma_F \) value \( (\sim 20 \mu \)m) has clearly increased, and such typical "high temperature" effects are further enhanced as the temperature increases. (Fig. 2).

III. PRELIMINARY MODEL FOR THE ETCHING OF PARTIALLY ANNEALED TRACKS.

III.1. Outline of the model. From the SAX results, we have developed the following model: i. The etching of the track stops when the distance, \( \lambda = 1/F(\Delta x \Delta D) \), between two successive \( \Delta x \Delta D \) exceeds a critical value \( \lambda_c \). This predominant role of the \( \Delta x \Delta D \) during the etching of a track is supported by our observation that etchable tracks are not formed in muscovite irradiated with light ions \( (Z < 9) \), which only generate PtD in this mineral; ii. \( \lambda_c \) depends on both the nature of the mineral and the previous thermal annealing history of the tracks. This last point can be understood by noting that the material separating 2 successive \( \Delta x \Delta C \) gets more difficult to etch out when the PtD have all been suppressed, and this in turn is equivalent to a decrease in the \( \lambda_c \) value; iii. For a given mineral, the "local" mean value of \( \lambda \), \( \lambda_c \), critically "depends on both the atomic number and the energy of the incident ions, and reaches a minimum value at an energy corresponding to the maximum of the dW/dX ionization rate curve. III.2. Verification of the model. By using these simple concepts, we have computed the theoretical dW/dL distributions expected for Fe-tracks in labradorite and muscovite. First we determine for these two different types of minerals the "room temperature" values of the \( \lambda \) and \( P(\Delta x \Delta D) \) parameters, by adjusting both the position, \( \Gamma_F \), and the half-width, \( \Gamma_{Fe}^c \), of the theoretical peak to the corresponding values experimentally measured for the non-annealed tracks. In labradorite we then easily reproduce the \( \Gamma_F \) and \( \Gamma_{Fe}^c \) values observed after either the low temperature or the high temperature annealing runs. In fact in our model the low temperature annealing simply decreases \( \lambda \) down to \( \lambda_c^* \), while still keeping \( P(\Delta x \Delta D) \) constant, and consequently only a shift in the peak position occurs (\( \Gamma_F \) keeps the non
an annealed value). In contrast after the high temperature annealing all the PtD have been removed and $\lambda^*$ is now constant, but $P(\text{ExD})$ starts to decrease in producing a marked broadening of the $dN/dL$ distribution. In our model the peculiar double peak structure observed after the low temperature annealing of 7 MeV/amu Fe-tracks simply reflects the etching of tracks intersecting the external surface of mica ("partial" tracks). Consequently, when $\lambda_c$ has decreased down to the $\lambda^*$ value, etching has first to progress into an ExD poor region where the probability of blocking the etching is high. Thus Fe-tracks can be suddenly cut after only a few microns, in being transferred into the "short-track" peak.

IV. THE CHEMICAL COMPOSITION OF VH AND VVH COSMIC RAY NUCLEI. IV.1. Iron group nuclei. First we note that several authors have argued that the broadening of the peak corresponding to the track lengths of VH nuclei ($20 \leq Z < 30$) in meteoric and lunar minerals can be used to infer the detailed chemical composition of such nuclei in the ancient cosmic rays. Unfortunately, our annealing studies show that such a broadening is quite compatible with a "thermal" broadening of the iron track distribution, corresponding to an $\lambda^*$ value which is about 3 times smaller than that observed for fresh iron tracks. We thus conclude that the relative abundance of heavy nuclei in the VH peak is difficult to infer from fossil track length studies. IV.2. VVH/VH ratios. We propose the following procedure in order to tentatively infer various VVH/VH ratios in the ancient galactic cosmic rays: i. From the position and width of the iron peak in a given mineral we can determine the maximum value, $P^{\text{max}}(\text{VH})$, of the $P(\text{ExD})$ function corresponding to the track producing VH nuclei. When the non-annealed $P^{\text{max}}(\text{VH})$ and $P^{\text{max}}(\text{VH})$ values have been measured for artificially accelerated nuclei, then the $P^{\text{max}}(\text{VH})/P^{\text{max}}(\text{VH})$ ratio yields the "degree" of natural thermal annealing from which the $P^{\text{max}}(\text{VH})/P^{\text{max}}(\text{VH})$ ratios can be evaluated. The $P^{\text{max}}(\text{VH})$ values are finally used to compute the partially annealed track length distributions expected for each group of VH nuclei in the fossil $dN/dL$ distribution. This in turn allows the direct determination of the right VVH/VH ratios.