469

# Laboratory for Space Physics

Department of Physics
Washington University/St. Louis

FACILITY FORM 602

| | |
|---|---|
| N70-30943<br>(ACCESSION NUMBER) | (THRU) |
| 18<br>(PAGES) | 1<br>(CODE) |
| CR-110442<br>(NASA CR OR TMX OR AD NUMBER) | 30<br>(CATEGORY) |

# On Some Annealing Characteristics of Heavy Ions Tracks in Silicate Minerals*

M. Maurette+
Laboratory for Space Physics
Washington University
St. Louis, Missouri 63130

---

*Work supported in part by NASA 9-8165 and McDonnell-Douglas Corporation Contract #Z80058T

+On leave from: Laboratoire de Spectrometrie de Masse du C.N.R.S., Boite Postale n$^{o}$1, 91 ORSAY, France.

SPP-19

## ABSTRACT

We have studied the thermal annealing of fossil tracks stored in hypersthene crystals of the Johnstown meteorite. We have observed that the annealing of the tracks depend on their initial length and therefore of the charge of the incident ions. The shortest tracks are annealed at a lower temperature than the longer ones. These long tracks in turn have been found to be less thermally stable on the average than fission fragment tracks.

After first pointing out some difficulties in the study of the annealing of heavy ion tracks in solids, we discuss the possibility of using thermal annealing experiment to identify the origins of the tracks stored in naturally or artificially irradiated silicate minerals.

## INTRODUCTION

The defects produced along the path of a heavy ion incident upon an insulator can delineate a particle track. The motion of such defects can be thermally activated, and as a consequence the track can be annealed if the solid is heated (1).

We are currently performing detailed annealing experiments on heavy ion tracks of different origins in both plastics and silicate materials with the following objectives :

a - To identify the origins of the fossil tracks in terrestrial and extra-terrestrial materials ;

b - To change the threshold for track registration in solid state track detectors ;

c - To identify the charge of the constituent nuclei of natural or artificial flux of nuclear particles.

More complete results including a more thorough discussion of their applications to lunar and meteoritic materials will be presented later(2). In this paper we want only to make the following points :

a - Thermal annealing curves can depend strongly on the methods used to observe the tracks ;

b - The thermal stability of nuclear particles tracks depends on the charge and energy of the incident particles ; the annealing can therefore be potentialy used to identify such an origin.

## EXPERIMENTAL TECHNIQUE AND RESULTS

### Meteoritic hypersthene

Maurette et al(3) have previously shown that hypersthene crystals in the Johnstown meteorite contain tracks produced by the VH nuclei ($Z > 23$) in the ancient galactic cosmic ray flux. The charge of the ions are correlated with their track lengths, the heaviest ions having the longest tracks.

We have studied the isochronal thermal annealing of such tracks by separating them into two groups according to their lengths : the shortest ones, having lengths smaller than 10 microns are essentially due to the iron group nuclei ; those longer than

30 microns correspond to ions with charges greater than 32.

We have first choosen several large hypersthene grains in the zone of the Johnstown meteorite containing the highest fossil track density ($\sim 5 \times 10^6$ tracks/cm$^2$). These crystals were then mounted in a disc of epoxy resin ; the disc was polished to produce a flat surface on the crystals and then etched 15 minutes in a hot concentrated solution of NaOH - details on the experimental technique can be found in reference 3. The densities $\rho_o(L < 10\mu)$ and $\rho_o(L > 30\mu)$ of the two groups of tracks were then measured for each crystal. The disc was repolished to remove the etched layer and the crystals annealed at different temperature ranging from 300° C to 700° C, during two hours. After the annealing the crystals were etched again and the values of $\rho_t(L < 10\mu)$ and $\rho_t(L > 30\mu)$ were measured. The annealing curves in figure 1 were obtained by plotting the ratio $\rho_t(L < 10\mu) / \rho_o(L < 10\mu)$ and $\rho_t(L > 30\mu) / \rho_o(L > 30\mu)$ as a function of the annealing temperature.

All the crystals were then heated for two hours at 700°C, remounted in a disc of epoxy resin, polished, irradiated with a known dose of Cf$^{252}$ fission fragments and then annealed at different temperatures and then etched. The densities of the fission fragment tracks were then measured and the ratio $\rho_t/\rho_o$ reported in figure 1.

It can be seen that there is a pronounced separation ($\sim$200° C) between the annealing curves of the two groups of fossil tracks, with the shorter tracks being less stable. The longer tracks in turn are less thermally stable than the fission fragment tracks.

## Muscovite Mica

In order to extend the scope of the discussion we show in Fig. 2 the annealing curves of α-interaction tracks [*] and fission fragment tracks measured in hornblende crystals by Crozaz et al (4). It can be seen that the α-interaction tracks are much less thermally stable than fission fragment tracks.

We have also studied the variations of the annealing curves of α-interaction tracks in mica as a function of the observation technique (ordinary transmission optical microscope, stereo-scan electron microscope) (Fig. 3).

## DISCUSSION OF THE RESULTS

### Dependence of the annealing curves on the technique used to reveal and observe the tracks

It can be seen from an examination of figures 2 and 3 that the position and the structure of an annealing curve depend on the technique of observing the tracks ; furthermore Huang et altri (5) and Widell (6) have shown that important changes in the annealing curves occur when the etching time is varied.

These dependences are particularly pronounced when the length of the tracks become smaller than two microns. Annealing curves must therefore be carefully defined in operational terms. A certain amount of subjective judgement is also present. This fact may make it difficult to compare annealing curves from different laboratories.

[*] The α-interaction tracks are produced by the recoiling nuclei produced during the elastic and inelastic interactions of α particles with the constituent atoms of the mica ; the most probable recoils have a $Z \sim 14$ and energies $\lesssim 0.5$ Mev/amu.

However such variabilities in the annealing curves don't constitute a trouble as long as the purpose of the annealing experiment is to identify the origin of tracks in a given material ; we can use the observation techniques which magnify the differences.

## Identification of the Origin of the tracks in naturally or artificially irradiated silicate minerals

A - Naturally irradiated samples :

The tracks stored in meteoritic or lunar minerals can have many different origins(6); for example they may be produced by the VH nuclei of the galactic or solar flare cosmic rays, by the fission fragments emitted during the spontaneous fission of extinct super heavy elements, or by the recoils produced by the interaction of the light ions of the cosmic ray flux with the constituent atoms of the minerals. Different criteria have been proposed to identify some of these origins(7) ; for example, we may measure the length and spatial distribution of the tracks and the variation of their density with the depth in the meteorite or in the grain. But this identification can be difficult under the following conditions :

a - When tracks of different origins have approximately the same length (e.g. : fission tracks and VH tracks) ;

b - When the tracks are so short that they are revealed only as shallow etch pits (in this case the length distribution* become very difficult to measure)

* However, Crozaz et al (2) have begun to measure the depth distribution of such shallow etch pits by using a stereo-scan electron microscope.

c - When the density is so high that it is not possible to measure the geometrical characteristics of individual tracks with an optical microscope.

The present results show that in such cases annealing experiments can be useful. For example, Fig. 1 shows that one can thermally differentiate fission fragment tracks from VH tracks. It is also possible to decrease the density of tracks in crystals in which this quantity is very large. The remaining tracks can then be studied with an optical microscope.

Finally we want to describe our use of annealing experiments in an attempt to determine the origin of the large density of etch canals recently observed in orthopyroxene crystals of the Kapoyeta gas rich meteorite by Pellas et al(8) and Lal et al (9). These etch pits are attributed by them to the tracks of the iron group nuclei of the solar flare cosmic rays. One of the difficulties in this identification is due to the very high value of the track density ($\sim 10^9$ tracks/cm$^2$) which prevents the study of the track distribution with an optical microscope. The observation of the tracks can only be made with a Stereo-Scan electron microscope after a very short etching time : therefore, only the most superficial part of each track can be observed (because the penetration of the electron beam in silicate materials $\lesssim$ 1 micron) and the measurement of the spatial and length distribution of the track cannot be made (see fig. 4). Consequently, we decided to anneal more than 20 Kapoyeta orthopyroxene crystals in order :

a - To decrease the density of the tracks in view of studying the geometrical characteristics of the partialy annealed tracks with an optical microscope ;

b - To verify that the etch canals distribution was annealed at a temperature corresponding to the annealing of the VH track distribution in meteoritic orthopyroxene crystals.

After 2 hours of annealing at 600° C the crystals were etched during a very short time and then studied with the Stereo-Scan electron microscope. A very striking change in the habit of the crystals was observed (see fig. 5) : the crystal is now divided in small blocks separated by materials containing very deep etch canals which cannot be tracks ; furthermore, the surface of the crystal is covered with a very thin and resistant reaction layer which prevent the observation of any canals similar to those represented in fig. 4, even if they still existed in the crystals.

Therefore this annealing experiment has not been useful to identify the origin of the etch canals observed in the crystals of the Kapoyeta mateorite. But we think that it was interesting to describe the thermally induced alteration of the crystals because such low temperature changes are unexpected in orthopyroxene crystals and therefore their studies should help to trace back the strange history of the Kapoyeta crystals.

B - Articicially irradiated samples :

Perelygin et al (10) have studied the ternary fission produced by argon ions in heavy elements using mica solid state track detectors. The tracks of the incident argon ionsin the mica were erased selectively by the use of annealing experiments, leaving only the tracks produced by the fission fragments. This experiment shows clearly that the heating of the detector before the etching changes the threshold of sensitivity for track registration in solid state track detectors. More results concerning such modifications will be reported later(2).

# BIBLIOGRAPHY

1 - R.L. Fleisher, P.B. Price, R.M. Walker.
Solid State Track Detectors : Applications to Nuclear Science and Geophysics, Ann. Rev. Nucl. Sci. 15, 1 (1965).

2 - G. Crozaz, M. Maurette, R.M. Walker.
On the annealing of heavy ion tracks in solid state track detectors : Applications to lunar and meteoritic materials.

3 - M. Maurette, P. Thro, R.M. Walker, R. Webbink.
Fossil tracks in meteorites and the chemical abundance and energy spectrum of extremely heavy cosmic rays, International Atomic Energy Agency Conference on Meteorites, Vienna (1968).

4 - G. Crozaz, M. Hair, M. Maurette, R.M. Walker.
Nuclear Interaction Tracks in minerals ; Applications to lunar and meteoritic studies, communication 3, Session VII, this conference.

5 - H. Huang, M. Maurette, R.M. Walker.
Observation of fossil α-particle recoil tracks and their implications for dating measurements, IAEA Symposium on Methods of Radioactive Dating, Monaco (1967).

6 - C.O. Widell.
The stability of patent tracks in mica, Session 1, this conference.

7 - R.M. Walker.
Fossil tracks : Studies in extra-terrestrial materials, Session VII, this conference.

8 - R. L. Fleischer, P. B. Price, R. M. Walker, M. Maurette.
Origins of particle tracks in meteorites, J. Geophys. Res. 72, 331 (1967)

9 - P. Pellas, G. Poupeau, J. C. Lorin, H. Reeves, J. Audouze.
Primitive low-energy particle irradiation of meteoritic crystals, preprint, to be submitted to Nature (1969).

10 - D. Lal, R. S. Rajan.
Observations relating to space irradiation of individual crystals of gas-rich meteorites, preprint, to be submitted to nature (1969)

11 - V. P. Perelygin, S. P. Tretiakova, N. H. Shadieva, A. H. Boos, R. Brandt.
On ternary fission produced in gold, bismuth, thorium and uranium with argon ions, preprint. Joint Institute for Nuclear Research, Dubna (1968).

# FIGURE CAPTIONS

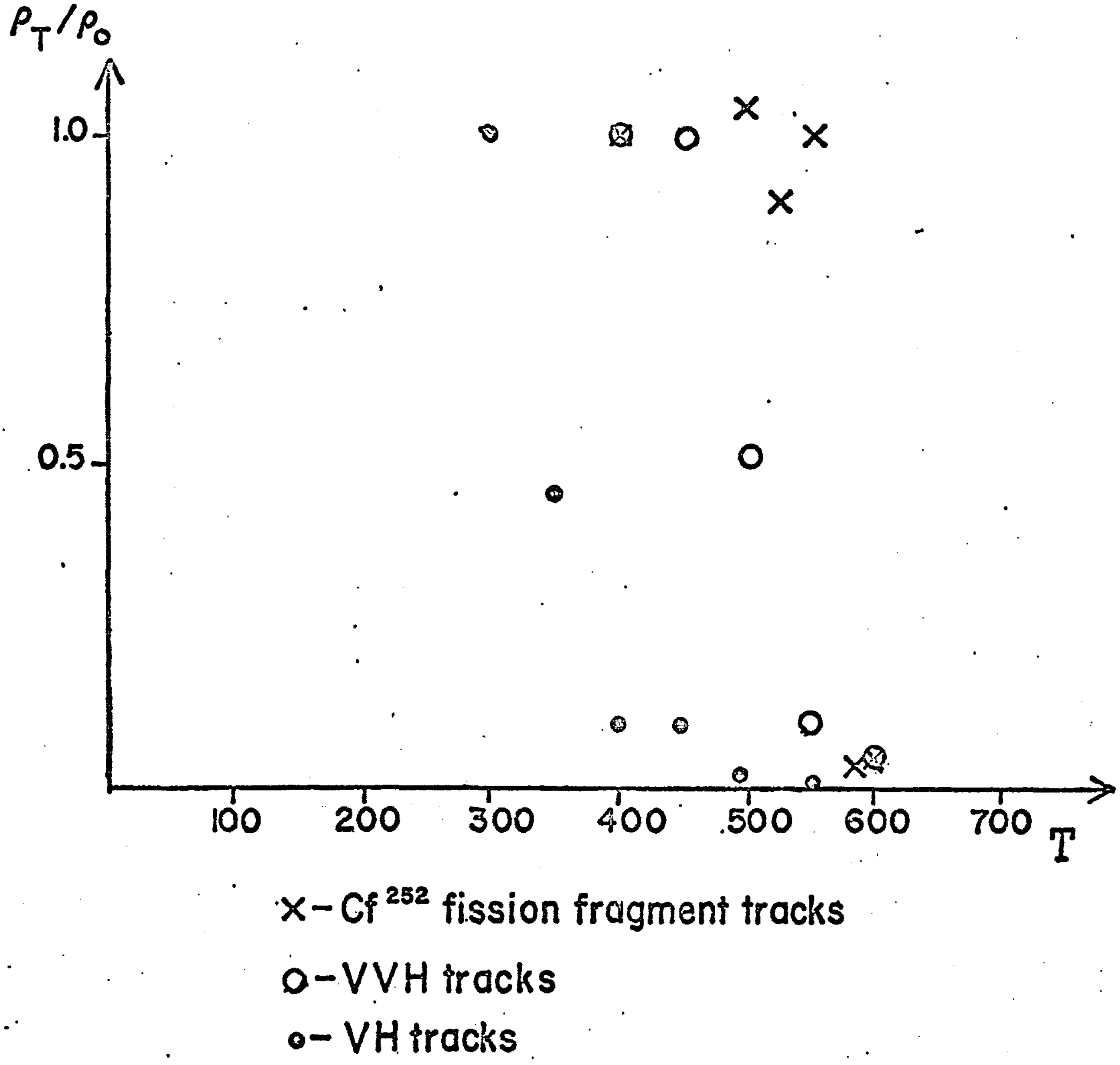
Fig. 1 Isochronal annealing curves of heavy ions tracks in hypersthene crystals of the Johnstown meteorite.

Fig. 2 Isochronal annealing curves of α-interaction tracks and fission fragment tracks in hornblende crystals.

Fig. 3 Effects of the technique of observation on the annealing curves of α-interaction tracks in muscovite mica.

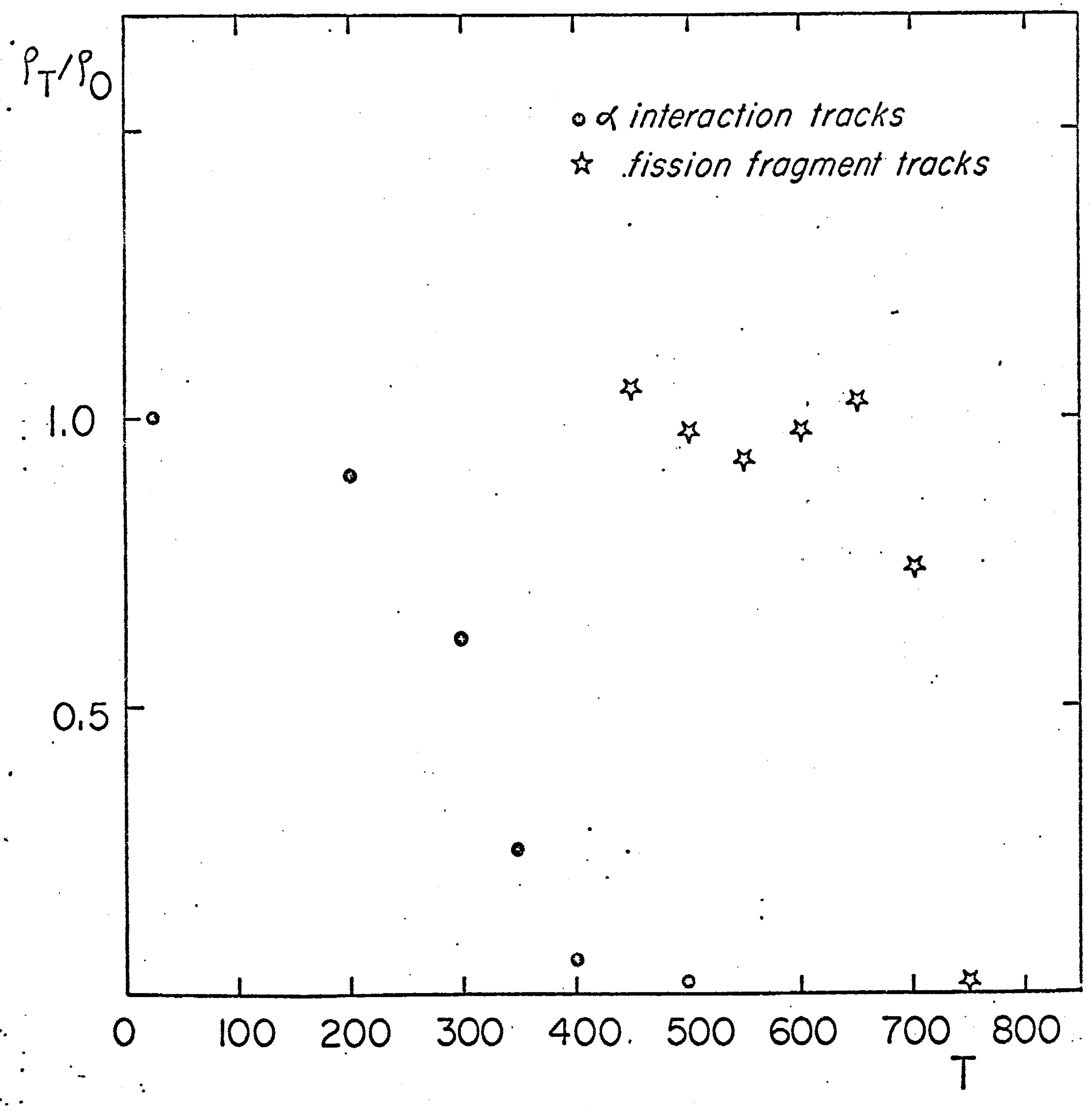
Fig. 4 Pipe-like etch pit distribution observed in an orthopyroxene crystal of the Kapoyeta meteorite. Stereo-scan electron microscope : magnification x 26,000.

Fig. 5 Crystal habit of orthopyroxene crystals of the Kapoyeta achondrite annealed 2 hours at 600° C and etched briefly. Stereo-scan electron microscope : magnifications 700 and 7,000.



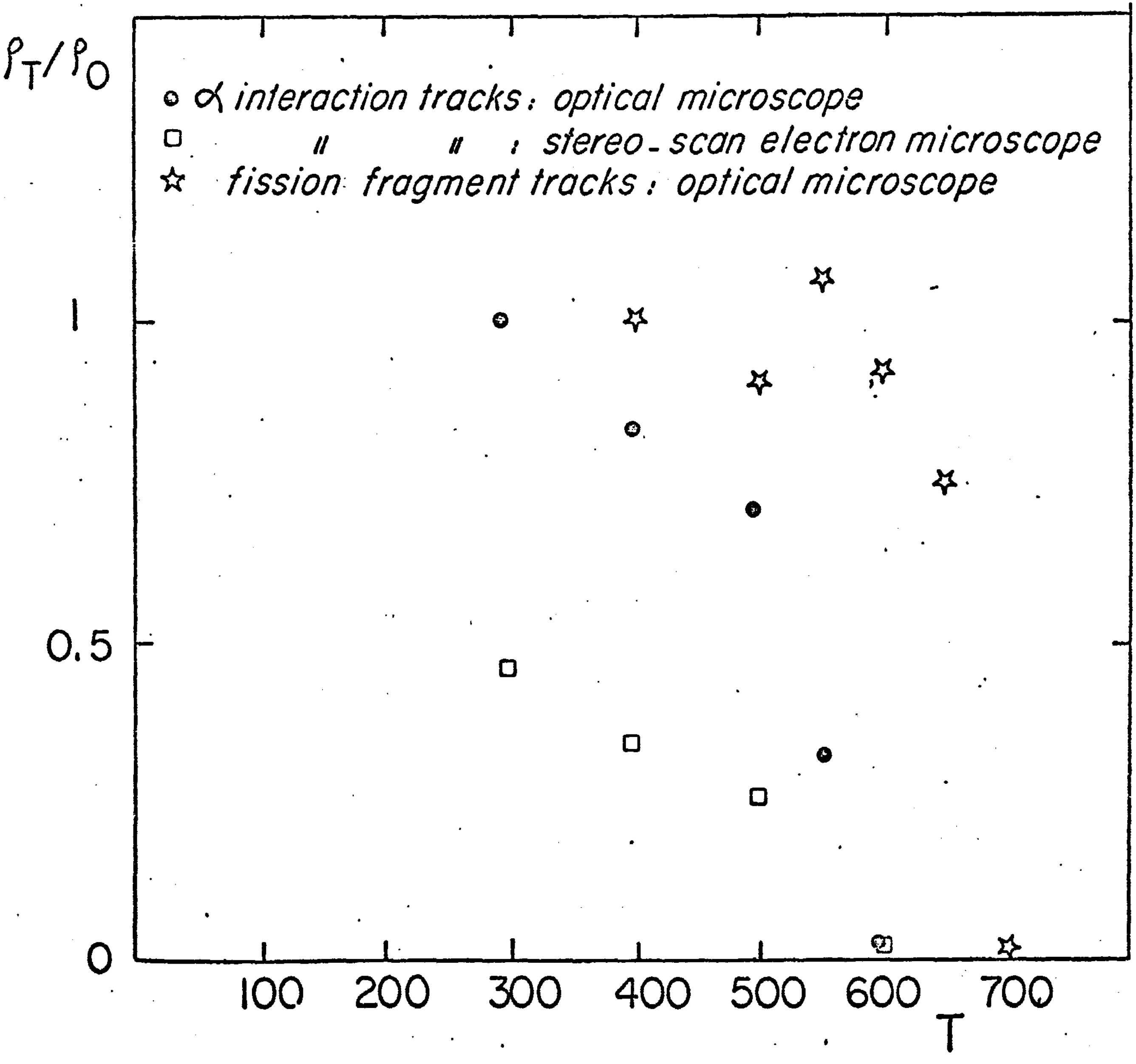
JOHNSTOWN HYPERSTHENE

— FIGURE 1. —



$\rho_T/\rho_0$
○ α interaction tracks
☆ fission fragment tracks
1.0
0.5
0
100
200
300
400
500
600
700
800
T

— Fig. 2 —



$\rho_T/\rho_0$
⊙ α interaction tracks : optical microscope
□ " " : stereo-scan electron microscope
☆ fission fragment tracks : optical microscope
1
0.5
0
100
200
300
400
500
600
700
T

— Fig. 3 —

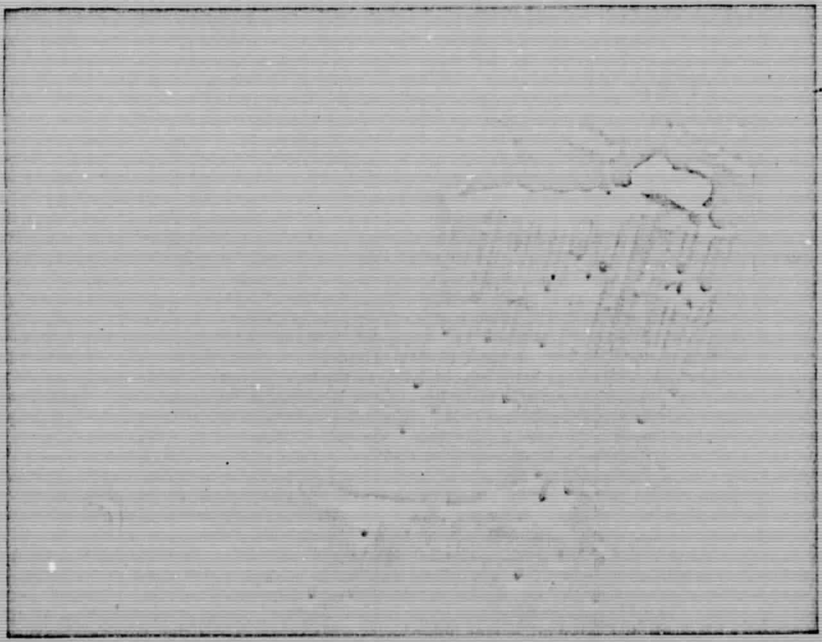


FIGURE 4

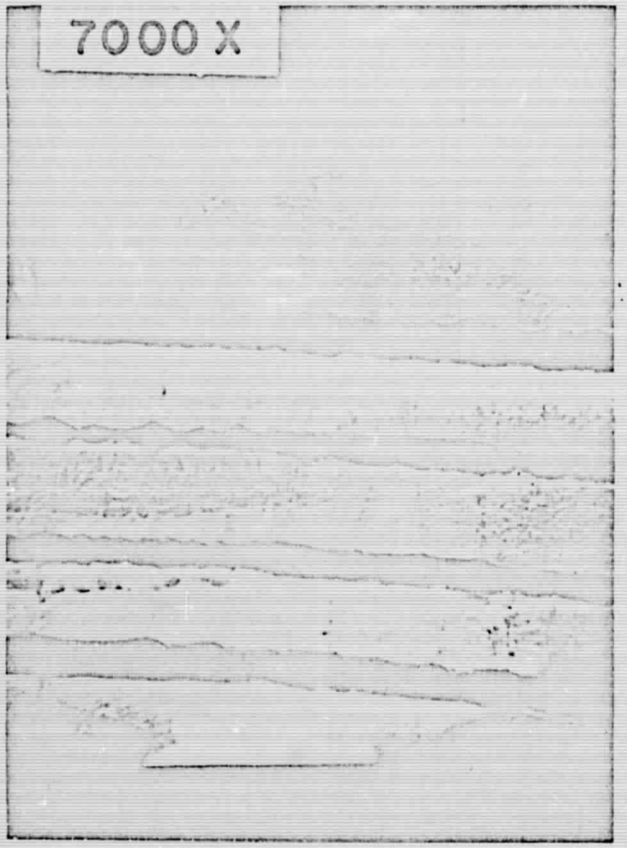


FIGURE 5