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Electron Microscope Investigation on the Nature of Tracks of Fission Products in Mica*

GUIDO BONFIGLIOLI, ANDREA FERRO, AND ADRIANA MOJONI
Istituto Elettrotecnico Nazionale Galileo Ferraris, Torino, Italy

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Specimens of biotite, muscovite, artificial fluorophlogopite, and muscovite annealed at 900°C were examined using an electron microscope after having been covered with a uranium layer and irradiated in a reactor. Tracks due to fission products appeared on every specimen except biotite, and the respective diameters were put into correspondence with the resistance of the various micas to thermal decomposition. The tracks were larger in muscovite (240 A), thinner in fluorophlogopite (150 A), and still more in dehydrated muscovite (115 A). The writers believe that the mechanism of damage has its origin in the heat released by the heavy ionizing particles. Peculiar recovery effects were often observed.

1. INTRODUCTION

THE electron microscope is the most suitable means to obtain direct information on the defects produced by fast particles irradiation on solids. Recent work on metals bombarded with fast neutrons has been performed by Hirsch[1] and collaborators, on graphite by Bollman,[2] on fission tracks in mica by Silk and Barnes,[3] and on evaporated uranium oxide by Noggle and Stigler.[4] Unpublished work[5] on tracks of fission fragments in muscovite performed through the same technique of Silk and Barnes had led the writers to the view that such tracks are essentially caused by thermal effects.

However, the mechanism of production of the observed tracks did not appear to be clarified completely, and it seemed that further studies dealing with different materials would be worthwhile. In particular from this viewpoint, materials with very similar crystal structures, but having different thermal decomposition points, look very suitable.

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[1] P. B. Hirsch and J. Silcox, Phil. Mag. 4, 1356 (1959).
[2] W. Bollman, Phil. Mag. 5, 621 (1960).
[3] E. C. H. Silk and R. S. Barnes, Phil. Mag. 4, 970 (1959).
[4] T. S. Noggle and J. O. Stigler, J. Appl. Phys. 31, 2199 (1960). We thankfully acknowledge the kindness of these authors, who let us see their paper prior to publication.
[5] G. Bonfiglioli, A. Ferro, and A. Mojoni, Final Report of the ARDC (U. S. A. F.) March, 1960.

2. EXPERIMENTAL

The materials chosen were biotite, muscovite, artificial fluorophlogopite, and muscovite which had been dehydrated by annealing one hour at 900°C.

The crystal structures of these micas are very similar. All are monoclinic and can be easily cleaved along the basal plane into extremely thin sheets.

However, they differ widely as to their resistance to thermal decomposition. Biotite loses crystallization water at about 500°C; muscovite does the same at 750°C, and fluorophlogopite does not decompose up to 1200°C. Muscovite dehydrated at 900°C decomposes further only at about 1400°C, where vitreous silicates are formed. In addition, at about 1100°C, some Al_2O_3 may be formed by prolonged annealing.[6]

Composite sandwiches of the micas and of natural uranium evaporated on high purity Al foils were prepared.[3] Micas were 30 μ thick, which corresponds roughly to the range of the fission fragments. A first series of specimens were irradiated in the swimming pool reactor of CENG (Grenoble, France) with an integrated flux of $1.1.10^{15}$ thermal neutrons/cm^2 and a second series in the swimming pool reactor of SORIN Corporation (Saluggia, Italy), with an almost identical flux.

A few hours after irradiation, the specimens were put

[6] Cf. R. Roy, J. Am. Chem. Soc. 32, 202 (1949); also W. Brindley, *X-ray Identification of Structure of Clay* (Miner Society, London, 1951).

FIG. 1. Fission tracks in muscovite (virgin); (magnification: 15 000 ×).

in a liquid nitrogen bath, where they were kept until observation (that is, some weeks at most). For the electron microscope examination the specimens were thinned by repeated cleavage until transparent. The final thickness, checked by shadow-casting, were between 1000 and 2000 A. All the pictures considered here were made with an Hitachi microscope (HU 11).[7] Over 400 pictures of different specimens were taken and the most significant ones are reproduced in Figs. 1, 2, and 3 (respectively: virgin muscovite, annealed muscovite, fluorophlogopite).

The magnification was carefully checked several times each day by a diffraction grating replica; the reproducibility—which is of primary concern in this work—was definitely better than 5%, even though the accuracy in the absolute calibration cannot be assumed to be as good as this. The electron magnification was 20 000.

To avoid excessive heating the pictures were taken with very low illumination and long time exposures, unless otherwise desired.

3. TRACK WIDTHS

The point of main concern was the measurement of the diameter of the tracks. In fact, we already suggested in our preliminary report[5] that tracks could represent thermal spikes caused by heat generated by the intense ionization in the wake of the fission fragments. Different interpretations have been suggested by Silk and Barnes,[3] such as Rutherford collisions or direct ionization effects.

More recently and independently, thermal spikes have also been taken into consideration by Noggle and Stigler[4] to account for the observed diameters of the tracks in irradiated uranium oxide. The sizes of the tracks observed in our experiments can rather well be accounted for on these grounds.

As is well known, viewing crystalline specimens with suitable objective apertures, we obtain local contrast (diffraction contrast) through local differences in the intensity of the Bragg diffracted beams. These considerations apply, for instance, to the observation of dislocations, whose strain field distorts the lattice. In the case of a thermal spike, the situation should be rather similar; in particular with mica, we think that the wake of the particle should consist of material which has been thermally altered and which therefore has a crystalline structure different from the adjacent region.

As a consequence, for limited portions of the range, the image to be expected—at least in a densely packed crystal—would be that of a cylinder, with a strong contrast in respect to the surroundings, its diameter being close to the actual diameter of the altered region.

Actually, in an open structure like mica, it cannot be surely stated that the altered region is cylindrical, since sensible effects of anisotropicity in the thermal diffusion could have occurred. Nevertheless, if we are only aiming at a rough evaluation, we may speak of the "characteristic dimension" of the altered region as of a diameter. This diameter can be calculated approximately from the stopping power of the fission fragments, taking into account average values of the various quantities involved.[8]

The data quoted show that the energy of the particle is, almost along its whole path, far in excess of the critical energy for prevailing displacement production

FIG. 2. Fission tracks in muscovite annealed (dehydrated) 1 hr at 900°C; (magnification: 15 000 ×).

[7] The examinations referred to in reference 5 were made with an old Siemens instrument. In addition to a great improvement of the resolving power, the presence of a double condenser strongly reduced heating.

[8] The average atomic weight of a typical mica is 20, its density 2.8 g/cm^3, its average atomic number 10. The mean value of the mass of a fission fragment is 117 and the mean initial energy about 78 Mev; hence its velocity is about 10^9 cm/sec. Accordingly, the particle is heavily ionized, with an initial charge of about $18e$ [A. H. Cottrell, Met. Rev. **1**, 479 (1956)]. The range of such a typical fragment in a typical mica can be estimated, from the data in argon [N. O. Larsen, Danske Videnskab. Selskab, Matt.-fys. Medd. **25**, 11 (1949); reproduced in Progr. Nuclear Phys. **2**, 146 (1952)] to be around 21 μ.

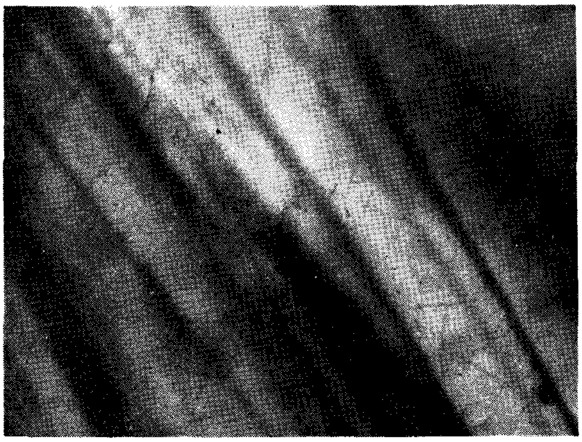


FIG. 3. Fission tracks in (artificial) fluorophlogopite; (magnification: 15 000 ×).

(cf. reference 10) (here less than 100 kev). The overwhelming majority of this energy is therefore spent in ionization. Of course in this order of approximation the energy lost per unit length in various micas can be considered the same—say 370 ev/A—the variation along the path being far greater in the same specimen.

Now, the diameter of a thermal spike which is assumed to be cylindrical can be easily evaluated, considering that the region where the energy is first released as ionization has a diameter which is small compared to that of the tracks.

Given a certain "critical temperature," let us call it T_c, it is easy to show that a cylinder of radius r exists, which marks the boundary between an external region which never reaches temperatures higher than T_c, and an internal region which at some instant is heated above T_c. This radius is

$$r = 2e^{-\frac{1}{2}}(Q/4cT_c)^{\frac{1}{2}},$$

where Q stands for the heat released per unit length and c for the heat capacity of the material.

It is interesting to note that the radius is independent of transport parameters, such as heat conductivity, and only dependent on structure parameters.

If the values of T_c are taken to be the thermal decomposition points which have been observed for bulk specimens of the different micas and if a mean stopping power of 370 ev/A is assumed for every mica, then the theoretical estimates reported in Table I, column A are obtained. Needless to say, this approach using a "mean stopping power," is very rough. On the other hand, it would be practically impossible to make a calculation which would take into account the various statistical distributions of the quantities involved in such a way as to be capable of accounting for the fairly broad distribution of diameters which is found experimentally on every picture. The above calculation is then justified insofar as only an order-of-magnitude value of the thermal spike diameter is sought.

As to the comparison with the experimental values,

TABLE I. Comparison of the calculated diameters of the tracks to the experimental values.

Material	Decomposition point	Diameter of the tracks (A) Calculated (from mean energy loss)	(B) Observed (mean)
Biotite	500° C	...	No tracks observed
Muscovite	700° C	120	240
Fluorophlogopite	1200° C	90	148
Muscovite heat treated 1 hr at 900° C	1400° C	80	115

let us remark that the statistical distribution of the widths of the tracks shown by the pictures arises from different causes. The prominent cause is due to the spectrum of initial energies and to the fact that different unknown portions of the ranges of the particles are recorded. Other minor causes are to be found in differences among tracks crossing regions of strong contrast, such as wide extinction contours; here the tracks are wider, by roughly 15%, than in clear regions. Lastly, differences in focusing can perhaps bring a further

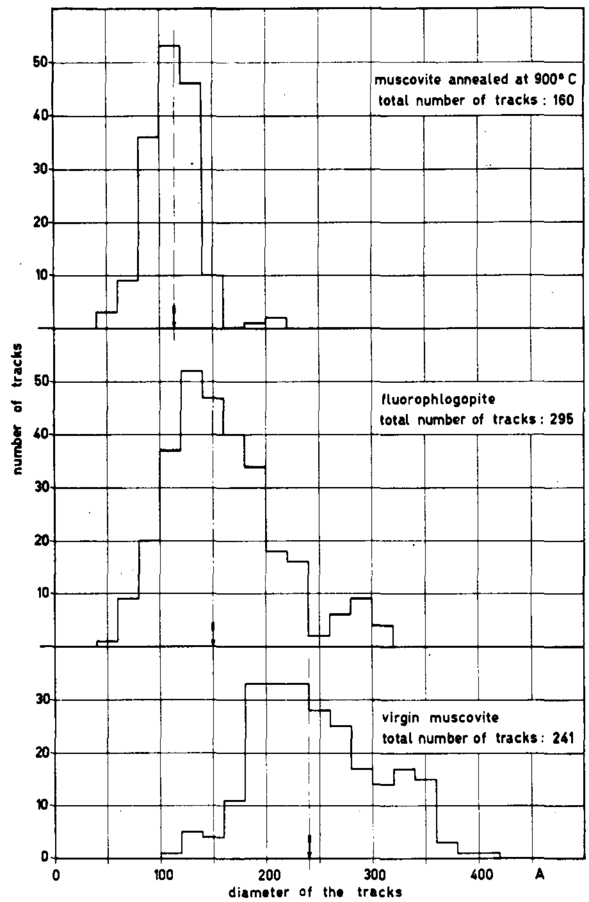


FIG. 4. Hystograms showing statistical distribution of the track diameters.

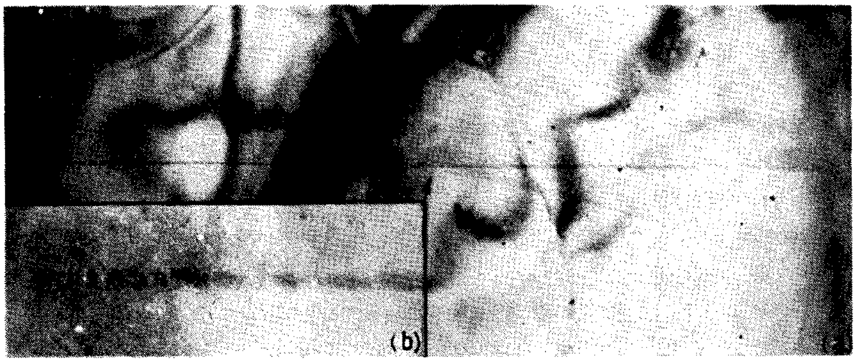


FIG. 5. (a) A long track (magnification 23 000 ×) in muscovite. (b) Enlarged detail of Fig. 5 (a), showing dotted region of the track (magnification 70 000 ×).

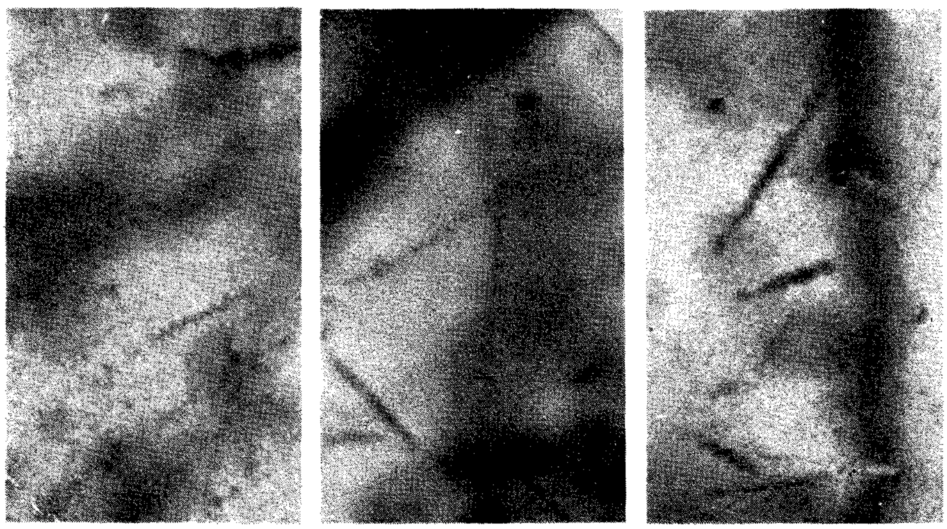
appreciable variation in the apparent diameter. To gather information about the effect of the significant variable alone (the nature of the material), we evaluated for each material a mean value of the tracks' widths obtained by averaging on the random variables, that is to say, by considering rather large groups of tracks on several pictures and for every material, whether in dark or in clear regions. The hystograms reproduced in Fig. 4 show that this procedure allows for a reasonable degree of confidence in the results. It can be seen that the mean values of the experimental distributions obtained by the indicated procedure are well separated for the three species of micas and are smaller the more resistant the mica is to thermal decomposition.

No doubt, the assumption of well-defined "critical temperatures" to account for thermal transients of some 10^{-11}-sec duration may appear objectionable. For a critical discussion of the theory of temperature spikes, the reader is referred to the article by Seitz and Koehler[9] and in particular to footnote 24, which precisely deals with the case of a fission fragment. In conclusion, the sensible differences in average width shown by tracks seen in different micas, and also the fact that the tracks are narrower in thermally more stable materials, strongly support the idea of their thermal origin.

4. OTHER EFFECTS

Some tracks (see Figs. 5 and 6) look dotted or dashed. We think this fact is essentially due to incipient recovery of the defects, as also indicated from the circumstance that fluorophlogopite and dehydrated mica do not show this phenomenon. Silk and Barnes[3] have interpreted the dots as due to Rutherford collisions which eject atoms from the mica lattice.

The effects indicating nuclear encounters may occasionally be detected. Moreover, it is indeed unquestionable that Rutherford collisions imparting to the struck nucleus an energy greater than the threshold for displacing it, occur roughly once every 100 atoms along the path of a fission particle.[10] However, the



(a) (b) (c)

FIG. 6. Helices and dotted tracks due to recovery in virgin muscovite (magnification 47 500 ×).

[9] F. Seitz and J. S. Koehler, in *Solid-State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1956) Vol. 2, p. 351 ff.
[10] A. H. Cottrell, Met. Rev. 1, 479 (1956).

energy spent in ionization is a thousand times greater and the Rutherford displacement encounters are superimposed onto a far heavier continuous ionization process, and therefore they do not seem to participate in the formation of the tracks.

Another explanation of tracks which can reasonably be ruled out is that of direct ionization effects. An estimate of the so-called "maximum radius" of Bohr's equation[11] gives for it a value of a few angstroms, as compared with the more than 100-A diameters of the tracks.

As previously stated, apparent changes in the widths of tracks occur when the tracks cross extinction contours [see Fig. 7 (a)]. We are inclined to regard these alterations as optical effects, which can in this case be considered as artifacts. A further example of this effect is found in Fig. 7 (b), where a small needle-shaped crystalline inclusion (perhaps Ti O_2) shows an apparent change in width as it crosses each of several extinction contours. However, intrinsic effects of a certain interest can be seen on some pictures.

Some tracks in virgin muscovite appear as distinctly resolved dots (see Figs. 5 (a) and (b) and Fig. 6); such tracks were not found in fluorophlogopite or in annealed mica, where the tracks always appeared as continuous thin black lines. We have interpreted the spottish aspect of these tracks as incipient recovery of the damaged region of the tracks under the electron beam of the microscope. Unfortunately, it has not been possible to check this by direct inspection, since all the pictures were taken with long exposure and low illumination in an effort to avoid excessive heating. It is interesting to note that the tracks shown in Figs. 6 (a), (b), and (c) look very much like the spiral dislocations which have been observed in quenched materials.[12] These features seem consistent with the fact that no tracks have been detected in biotite, which is the least heat-resistant mica examined. The preparation and irradiation of the biotite specimens was performed in exactly the same way as for other micas. It is therefore reasonable to suspect that in this material tracks have recovered during the irradiation or, more likely, under the microscope beam, even at the low illumination used.

(a)

(b)

FIG. 7. (a) A clear effect of modulation of darkening at the crossing of an extinction contour (magnification 42 000 ×); (b) effect of darkening modulation of a spurious needle-shaped crystal (magnification 15 000 ×).

CONCLUSION

The results of the investigation of fission fragment tracks in biotite, in muscovite dehydrated at 900°C, and in (artificial) fluorophlogopite indicate that the tracks are essentially thermal spikes whose widths are in rather good agreement with those calculated from the decomposition temperatures. The tracks are definitely thinner in fluorophlogopite and dehydrated muscovite than in virgin muscovite. Important recovery effects under the beam are observed in the latter. In biotite, where the recovery effects are likely to be more intense, no tracks have been observed.

ACKNOWLEDGMENTS

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[11] E. Fermi, *Nuclear Physics* (University of Chicago Press, Chicago, Illinois, 1950) p. 27.

[12] See, for example, K. H. Westmacott, D. Hull, R. S. Barnes, and R. E. Smallman, Phil. Mag. 4, 1089 (1959).