Nuclear-Particle-Track Identification in Inorganic Solids

R. L. FLEISCHER, P. B. PRICE, AND R. T. WOODS
General Electric Research and Development Center, Schenectady, New York 12301
(Received 10 July 1969)

The unique feature of track-recording plastics which allows them to be used to identify particles is that the chemical rate of attack V_T along a track is an increasing function of ionization rate. This same characteristic is shown to extend to inorganic dielectric solids. The relation between V_T and ionization rate has been measured for five different glasses, using fission fragments of different energies. The variation of V_T is least for ordinary lime glass and much the greatest for a phosphate glass, suggesting that the latter should be useful in resolving atomic numbers and energies of fission fragments and other very heavy, energetic nuclei. Equations are derived that allow the shapes of etched tracks to be calculated as a function of etching time and particle ionization rate. The calculations agree well with recent experimental results.

1. INTRODUCTION

PARTICLE identification by measurements on etched tracks in solids is possible because the chemical attack rate V_T along a particle track is a unique, measurable function of the localized damage caused by a particle.[1–3] This function has been inferred in a number of plastics,[1,3] and sheets of these plastics have been used to demonstrate that identification of neighboring elements is possible for particles of atomic number Z as high as 28 (Ref. 3) and that under proper circumstances neighboring isotopes as heavy as B^{10} and B^{11} may be clearly distinguished.[1]

The purpose of this work is to demonstrate that in inorganic glasses, V_T is also an increasing function of ionization rate and to indicate how and where such glasses might be used for particle identification.

One notable difference between the inorganic and the organic solids in their characteristics for particle track registration is that the ionization threshold is considerably higher for the inorganic solids—glasses and crystals.[4] The result is that while heavy ion accelerators are able to supply a variety of particles of known mass, charge, and energy necessary for calibration of the registration threshold of plastics, for the less sensitive minerals and glasses the choice is more limited. Particles that can presently be used include fission fragments and heavy ions such as I and Br, which are available at low energies per nucleon from certain tandem van der Graff accelerators.[5] In this work, we use fission fragments of various energies from a Cf^{252} source as a means of displaying the widely differing registration characteristics of a group of glasses.

2. RELATION OF CONE ANGLE TO ETCHING RATIO

Particle identification by solid-state nuclear-track detectors depends on the facts that the velocity of etching V_T along the damaged material comprising a particle track is greater than the general attack rate V_G of the undamaged material and is a unique function of the charge, mass, and energy of a particle. These facts lead to a geometry of etched tracks that can be understood with the aid of Fig. 1. Assume for simplicity [as sketched in Fig. 1(a)] that V_G is isotropic and that V_T is constant over the length $V_T t$ etched in time t. A surface layer of thickness $V_G t$ is etched away, and a conical etched track is produced of cone angle $\theta=\arcsin(V_G/V_T)$ and diameter D which can readily be shown to be $2V_G t[(V_T-V_G)/(V_T+V_G)]^{1/2}$ for a track normal to the etched surface. In short, both the cone angle and the diameter are functions of V_T, which in turn is a function of ionization (previously measured for plastics and shown here to apply to glasses also). Hence, knowledge of either θ or D makes particle identification possible just as the more direct quantity

$$L\left(\equiv\int_0^t V_T dt\right)$$

has been used by methods we have previously described.[1–3,6]

If the angle of the track to the surface φ is less than θ, then the rate of preferential etching normal to the

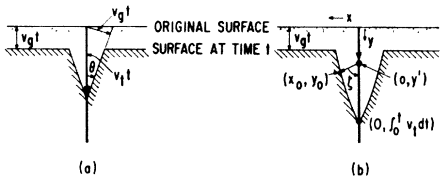

FIG. 1. Track geometry: (a) With V_T constant, the cone angle is $\sin^{-1}(V_G/V_t)$. (b) With V_T decreasing as etching continues, the shape of the etched track is more complicated.

[1] P. B. Price, R. L. Fleischer, D. D. Peterson, C. O'Ceallaigh, D. O'Sullivan, and A. Thompson, Phys. Rev. **164**, 1618 (1967).
[2] P. B. Price, R. L. Fleischer, D. D. Peterson, C. O'Ceallaigh, D. O'Sullivan, and A. Thompson, Can. J. Phys. **46**, S1149 (1968).
[3] P. B. Price, R. L. Fleischer, D. D. Peterson, C. O'Ceallaigh, D. O'Sullivan, and A. Thompson, Phys. Rev. Letters **21**, 630 (1968).
[4] R. L. Fleischer, P. B. Price, R. M. Walker, and E. L. Hubbard, Phys. Rev. **156**, 353 (1967).
[5] C. D. Moak, J. H. Neiler, H. W. Schmitt, F. J. Walter, and G. F. Wells, Rev. Sci. Instr. **34**, 853 (1963).
[6] P. B. Price, R. L. Fleischer, and D. D. Peterson, in *Reactivity of Solids*, edited by J. W. Mitchell, R. C. DeVries, R. W. Roberts, and P. Cannon (Wiley-Interscience, Inc., New York, 1969), p. 735.

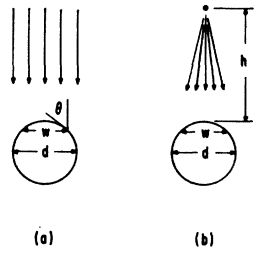

FIG. 2. Measurement of the critical angle θ for track etching: (a) Using a collimated beam of particles, tracks may be revealed over a chord width w on a cylinder (or sphere), and $\theta=\cos^{-1}(w/d)$. (b) With a point source,

$$\theta=\cos^{-1}(w/d)-\tan^{-1}\{w/[2h+d+(d^2-w^2)]\}^{1/2}.$$

surface, $V_T\sin\varphi$, is less than the rate V_G at which material is dissolved away from the surface, so that no track is revealed. This purely geometric result, described in more detail previously,[7] means that the particular value of the angle φ_c at which tracks cease to be revealed is equal to θ, and hence the etching ratio (V_T/V_G) is given by $1/\sin\varphi_c$. By measuring φ_c for particles incident on a surface at different ionization levels J, a relation between (V_T/V_G) and J may be constructed, the critical relation needed for particle identification.

3. EXPERIMENTAL VARIATION OF ETCHING RATIO WITH IONIZATION

We have utilized the simple geometry sketched in Fig. 2 to measure φ_c and deduce $\theta\equiv\arcsin(V_G/V_T)$. In the simplest case [Fig. 2(a)], a rod or cylinder of detector of diameter d is exposed to a point source of particles (in this case from a californium-252 source). Then the sample is etched and the chord width w is measured over tracks which are revealed by etching. θ is given by $\arccos(w/d)$. In the actual geometry used [Fig. 2(b)] a point source is spaced a distance h from the detector. The geometry in this case is such that

$$\theta=\varphi_c=\arccos(w/d)-\arctan\{w/[2h+d+(d^2-w^2)^{1/2}]\}.$$

Since in these experiments $h=2.6$ cm and d is typically 0.6 mm, the maximum value of the second term in θ is ≤ 0.1 radian. This term, although significant, is normally small except for the phosphate glasses where θ can also be small.

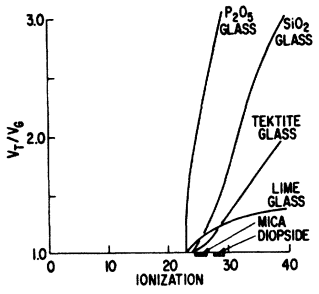

FIG. 3. Etching ratio V_T/V_G versus ionization rate J (relative values) for a group of glasses.

[7] R. L. Fleischer and P. B. Price, J. Geophys. Res. **69**, 331 (1964).

TABLE I. Relation between the etching ratio (V_T/V_G) and thickness of absorbing material for various glasses.

Absorber thickness (mg/cm²)	Etching ratio (V_T/V_G) [a] P_2O_5 glass	SiO_2 glass	Tektite glass	Obsidian	Lime glass
0.004	57 (±10)				1.381
0.088	$14.3\left(^{+\infty}_{-8.5}\right)$	2.96	1.79, 1.97	2.29	
0.307		2.64, 2.59			
0.526		2.78			1.363
1.038	$8.8\left(^{+\infty}_{-4.6}\right)$	1.98	1.46, 1.60		1.286
1.257	$5.3\left(^{+11.5}_{-3.4}\right)$	1.6			
1.476		1.72	1.24, 1.33		1.248
1.695	$2.14\left(^{+0.56}_{-0.38}\right)$	1.33	1.06, 1.11	1.125	1.15
1.914	$1.86\left(^{+0.24}_{-0.30}\right)$	1.0	1.022		1.086
1.988	1.0	1.0			1.0

[a] In mica, tracks are seen after 1.607 mg/cm² but not after 1.914; in diopside, tracks are seen after 1.476 mg/cm² but not after 1.607.

Tubes of lime glass[8] and rods of fused quartz were obtained and rods were made of a phosphate glass,[9] a synthetic tektite glass,[10] and a remelted obsidian.[11] The critical angle φ_c was measured either as described with reference to Fig. 2 or by directly rotating the sample through an angle $\pi-2\varphi_c$ so as to view normal to the surface at the two edges of the region in which tracks are revealed by etching. Cf^{252} fission fragments were used either in a vacuum of 1 mm or of 2 cm [the latter equivalent to 0.088 mg/cm² of air in our geometry of Fig. 2(b)]. Polycarbonate sheets of 0.219 and 0.950 mg/cm² were used to slow down the fission fragments by various amounts. Table I lists the thicknesses of absorbing material (foils plus air) for each experiment along with the etching ratios deduced. We used the range-energy data of Kahn and Forgue[12] to find the energies of the median light and median heavy fission fragments, and from the ionization curves deduced earlier for minerals[13] we calculated values of the ionization rate J in the glass. In each case, the J values for the more heavily ionizing of the two fission fragment mass groups were used. Figure 3 gives the resulting curves of (V_T/V_G) versus J. For reference the thresholds of muscovite mica and the same diopside reported earlier[13] are given, *threshold* being defined as the value of ionization at which fission fragments at normal incidence cease to be revealed by etching.

It is worth noting that all of the glasses have thresholds at least as low as the mica and that two, the lime

[8] Composition given by R. J. Charles, Amer. Ceram. Soc. **45**, 105 (1962).

[9] Glass No. 1457 of composition (wt %) P_2O_5, 63.00; BaO, 11.03; Ag_2O, 9.30; K_2O, 8.47; Al_2O_3, 8.20.

[10] Supplied by D. B. Chapman; made by Corning; wt % composition: SiO_2, 71.0; Al_2O_3, 13.6; FeO, 4.60; MgO, 2.5; CaO, 3.3; Na_2O, 1.5; K_2O, 2.5; TiO_2, 0.8; MnO, 0.1.

[11] Supplied by W. W. Patton; used as a particle detector in a previous study: R. L. Fleischer, P. B. Price, and R. T. Woods, Phys. Rev. **184**, 1398 (1969).

[12] S. Kahn and V. Forgue, Phys. Rev. **163**, 290 (1967).

[13] P. B. Price, R. L. Fleischer, and C. D. Moak, Phys. Rev. **167**, 277 (1968).

glass and the tektite glass are more sensitive than the mica.[14] In all cases the track etching rate rises monotonically with ionization, but the rate of increase varies widely from one to another and is most rapid for the phosphate glass.

4. TRACK DIAMETER AS FUNCTION OF V_T

As we noted earlier, track length, diameter, and cone angle are all functions of V_T/V_G, hence are related to the ionization, and therefore possible particle identification. Where values of θ are small, tracks are long and narrow, and consequently L is the most sensitive and easily measured parameter and the one we have used in previous studies.[1–3] Where θ values are large (as in many glasses) and in cases where particles enter at right angles to the detector surface, D becomes the preferred parameter. It is of interest to know how D will vary with etching time and with the applicable relation $V_T(y)$ for the variation of track etching rate with position along the track.

We now proceed to show how $D(t,V_T(y))$ can be calculated for tracks at normal incidence, referring to Fig. 1(b). First, using the coordinate system indicated there we calculate the shape of the etch pit $y_0(x_0)$ for an arbitrary $V_T(y)$ describing the attack rate for a track along the y axis. Twice the value of x_0 when $y_0=V_G t$ gives the diameter as a function of time. The etching time to reach the point (x_0,y_0) is given by

$$t=\int_0^{y'}\frac{dy}{V_T(y)}+\frac{[(y_0-y')^2+x_0^2]^{1/2}}{V_G}, \tag{1}$$

which is composed of two terms, the time to etch along the track from (0,0) to (0,y') given by

$$\int_0^{y'}\frac{dy}{V_T(y)},$$

and the time to etch from (0,y') to (x_0,y_0) given by $[(y_0-y')^2+x_0^2]^{1/2}/V_G$. Since the actual path from (0,0) to (x_0,y_0) is that corresponding to least time, we take the derivative of time with respect to y' and set it equal to zero, giving the relation

$$y'=y_0-x_0/[(V_T(y')/V_G)^2-1]^{1/2}, \tag{2}$$

which, together with Eq. (1), defines x_0 and y_0 for a given t, y', and $V_T(y)$. By noting that the angle ζ in Fig. 1(b) is arcctn$[(y_0-y')/x_0]$, we can write (1) and

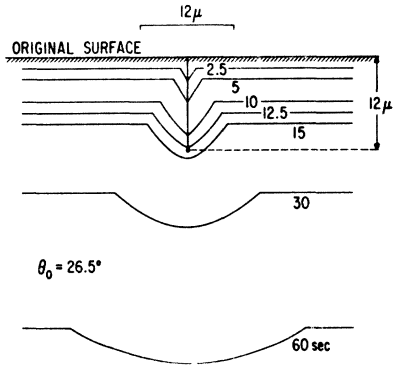

FIG. 4. Etching of a fission track in a glass of initial cone angle 26.5°. For this calculation it was assumed that $V_g=0.58$ μ/sec, $V_T=1.36-0.0657y'$ μ/sec, and $R=12$ μ. The initial cone angle is appropriate to obsidians.

(2) in a more useful parametric form in y' as follows:

$$x_0=[(y_0-y')^2+x_0^2]^{1/2}\sin(\zeta),$$
$$y_0=y'+[(y_0-y')^2+x_0^2]^{1/2}\cos(\zeta),$$

which can be rewritten

$$x_0=V_g\left(t-\int_0^{y'}\frac{dy}{V_t(y)}\right)\left(1-\frac{V_g^2}{V_t(y')^2}\right)^{1/2}, \tag{3}$$

$$y_0=y'+\frac{V_g^2}{V_t(y')}\left(t-\int_0^{y'}\frac{dy}{V_t(y)}\right). \tag{4}$$

These equations are valid for all y' and t.

Although in general these two equations can not be put into a closed expression for $D(t)$, the procedure for

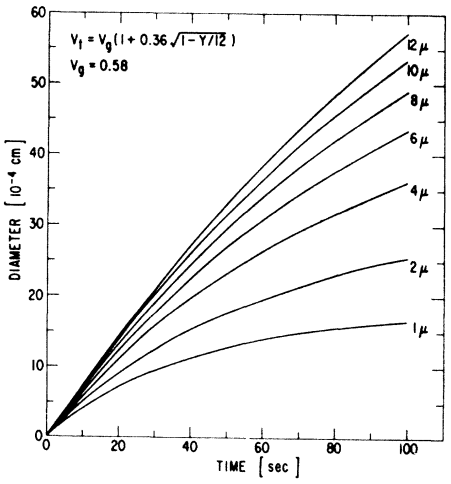

FIG. 5. Diameter versus time for etched tracks of different lengths in glass. In this calculation, we assumed $V_T=V_G[1+0.36\times(1-y'/12)^{1/2}]$, where y' is the distance along the etchable particle range, and starting values of y' of 0, 2, 4, 6, 8, 10, and 11 were used corresponding to fission track lengths of 12, 10, 8, 6, 4, 2, and 1 μ. $V_g=0.58$ μ/sec. The conditions considered are appropriate to ordinary soda glass (microscope slides, etc.).

[14] In previous reports, we have described mica as generally *more* sensitive than the glasses [R. L. Fleischer, P. B. Price, and R. M. Walker, Science **149**, 383 (1965); Ann. Rev. Nucl. Sci. **15**, 1 (1965)] as a result of calibration experiments in which particles were incident at 30° to the surfaces. Since only particle tracks for which $V_T/V_G>2$ were therefore etched in the glasses, they appeared less sensitive.

calculating $D(t)$ is straightforward: Values of y' are substituted in (4) until that value of y' for which $y_0 = V_G t$ is obtained; this value of y' in Eq. (3) gives $\frac{1}{2}D$ directly. It can be readily confirmed that for the simple case, $V_T = \text{const}$, the result quoted in Sec. 2 obtains.[15]

As an example of the use of these equations, Fig. 4 shows the profiles calculated for an etched track for various etching times and a specific, simple choice of V_T (decreasing linearly to V_G to the end of the track). The track remains pointed until its end is reached and then rounds out. The diameter increases monotonically at a decreasing rate. Although larger diameter differences are obtained at long etching times, the unique, pointed character of the tracks is lost and they become difficult to discriminate from background pits.

5. COMPARISON WITH EXPERIMENT

It has been shown by Somogyi[16] for plastics and Fiedler and Höppner[17] for glasses that the diameters of etched tracks are a function of the energies of the track-forming particles. In both cases the results derive simply from the variation of V_T along a track (as we showed in the plastics[1] and have shown here for glasses) and from the differing ranges of different particles. Since Fiedler and Höppner have measured diameters of tracks in glass as a function of time for Cf^{252} fission fragments, their work provides a useful opportunity to check the calculations outlined here. In Fig. 5, we have used an empirical fit to the data in Table I for lime glass to calculate diameter-time relations for fission fragments slowed down to lower energies so as to have residual ranges of 12, 10, 8, 6, 4, 2, and 1 μ. Figure 6 is a comparison of the diameters so calculated with those measured by Fiedler and Höppner. The agreement is excellent, well within the probable errors of the measurements, lending strong support to the description of the etching process given by Eqs. (3) and (4).

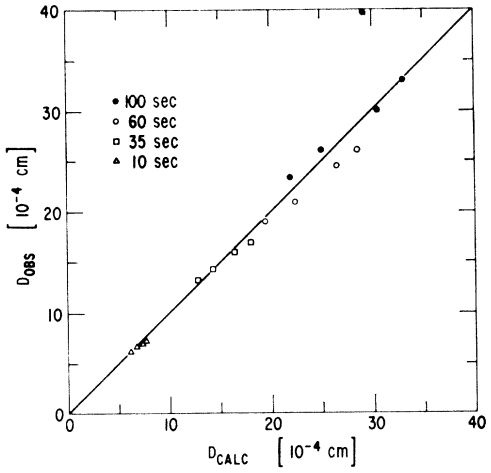

FIG. 6. Comparison of calculated diameters with those measured by Fiedler and Höppner for fission fragments of various energies in glass. We assumed $V_T = 0.43[1+0.36(1-y'/12)^{1/2}]$ μ/sec etching rate along the track of full energy, light, Cf^{252} fission fragments. For each time the points correspond to fragments of energies 80, 65, 40, and 34 MeV.

6. PARTICLE IDENTIFICATION

Particles may be identified if their energy (or residual range) and energy loss rate (or ionization rate) are known at any position along their path. It follows that for known particles, curves such as those given in Fig. 5 allow the energy to be determined, as has been shown empirically previously.[16,17] Where both the particle and its energy are unknown, they can be determined by making length measurements (as before[1-3,6]) or diameter measurements at two or more points along the trajectory or at two or more etching times. The energy is related to range through an appropriate range-energy relation (e.g., see Ref. 18).

Figure 7 demonstrates the decreasing ionization rate along fission tracks. A sheet of SiO_2 glass thinner than the range of fission fragments was irradiated and etched, giving rise to cone-shaped tracks on both surfaces, with obvious decreases in diameters on the lower surface resulting from the decreased ionization rate. The continuations of the particle tracks into a second piece of glass have also been observed, so that it is certain that these particles crossed the sheet shown in Fig. 7. Cone-length measurements from the glass in Fig. 7, together with measurements of track range in the second piece of glass, provide for each event two independent values of ionization rate at known residual range and thus provide two independent ways of identifying each particle.

For a detector having small cone angles (such as the phosphate glass[9] used here), Fig. 8 shows that diameter measurements after short etching times

[15] These calculations are based on the assumption that any accelerated *radial* attack outward from the core of the track may be neglected. This assumption is supported by calculations [R. L. Fleischer, P. B. Price, and R. M. Walker, J. Appl. Phys. **36**, 3645 (1965); R. Katz and E. J. Kobetich, Phys. Rev. **170**, 401 (1968)] which show that the region of most intense ionization and energy deposit at a track is strongly localized over the first few tens of Å. Experiments on glasses [R. L. Fleischer and P. B. Price, J. Appl. Phys. **34**, 2903 (1963)], crystals [P. B. Price and R. M. Walker, J. Appl. Phys. **33**, 3407 (1962); C. P. Bean, M. V. Doyle, and G. Entine, General Electric Research and Development Center Report No. 69-C-043, 1969 (unpublished)], and even plastics [W. DeSorbo and W. A. Healy (to be published)] make it clear that radial effects do not exist at distances of >200 Å from the center of a particle track and hence are quite properly neglected in considering effects on an optical scale.

[16] G. Somogyi, Nucl. Instr. Methods **42**, 312 (1966).

[17] G. Fiedler and U. Höppner, in *The Proceedings of the International Topical Conference on Nuclear Track Registration in Insulating Solids and Applications, Clermont-Ferrand* (University of Clermont-Ferrand Press, Clermont-Ferrand, France, 1969), Vol. 1, IV-35.

[18] R. P. Henke and E. V. Benton, U. S. Naval Radiological Defense Laboratory, San Francisco, Report No. USNRLL-TR-1102, 1966 (unpublished).

reveal only the value of V_g and hence contain no specific information on particle identification. Only after etching fission tracks long enough that the effects of the larger cone angles which obtain at low residual range are felt do the curves separate. A more useful procedure for this case is to measure V_T directly.

This can be done even for short tracks in a simple manner by making sandwich structures,[19] consisting of alternate layers of detector and metal film, which can be etched through the detector layers to the metal layers which are dissolved by the same etchant. The diameters of the holes in the opaque metal films can be related to the track etching rates (averaged over the individual thickness of the detector films).

7. DISCUSSION

It should be clear that the inorganic glasses are detectors which are capable of particle identification by the same principle that allows identification in plastics. There are two striking differences: First, the glasses are less sensitive and hence do not respond to the passage of many of the particles that will create tracks in plastics, and for other particles will record their tracks only over a smaller fraction of the range. Second, the glasses are less affected by elevated temperatures and by background effects such as the ambient atmosphere or the presence of lightly ionizing and electromagnetic radiation. The glasses are therefore especially suited to observing very heavy cosmic rays in long space exposures without the need for rigid control of the ambient conditions. In studies involving heavy element synthesis, glass detectors would also be appropriate for seeking occasional fission fragments from super-heavy trans-actinide elements in the midst of a copious background of ordinary fission fragments. Similarly, glass detectors should be useful in detecting any short-range heavily ionizing particles in the midst of a background of more lightly ionizing radiation for example, spallation recoil nuclei and fission fragments.

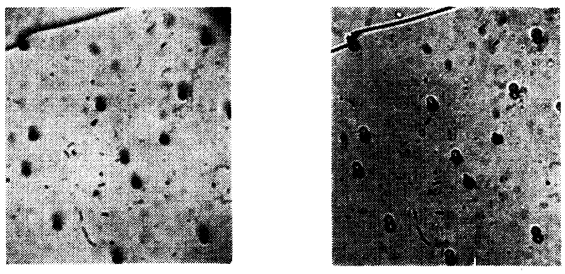

FIG. 7. Decrease in etching rate with slowing down of Cf^{252} fission fragments. Left: etched tracks on upper surface of thin sheets of SiO_2 glass viewed in reflected light. Right: the same etched tracks on lower surface viewed in transmitted light. Decreases in ionization rate during slowing down are revealed by smaller diameters of the pits on the lower surface.

[19] R. L. Fleischer, P. B. Price, and J. R. M. Viertl (to be published).

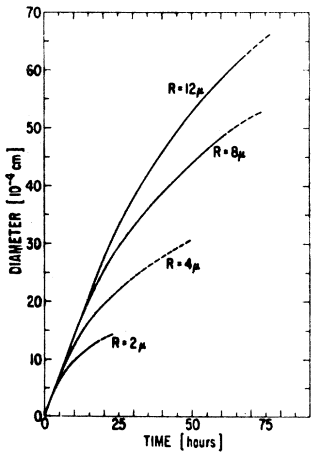

FIG. 8. Diameter versus time for etched fission tracks of different lengths in a glass with a small cone angle. In this calculation, we assumed $V_T = V_G[1+14(1-y'/12)^{1/2}]$, where y' is the distance along the etchable particle range, and starting values of y' of 0, 4, 8, and 10 were used corresponding to fission track lengths (R) of 12, 8, 4, and 2 μ. $V_g = 0.7$ μ/h. The conditions are appropriate to P_2O_5 glass (Ref. 9). Because $V_T \gg V_g$, the initial increase in diameter is given by V_g, so that difference in diameters are imperceptible. Either length measurements for short times or diameter measurements for long times are necessary to discriminate between different particles.

8. CONCLUSIONS

(a) In glasses as in plastics, etching rates along particle tracks are an increasing function of ionization rate and thus make possible particle identification.

(b) Etching rates vary with ionization level in widely different degrees for different glasses. Phosphate glass has the highest resolution; that is, it has the greatest change in etching ratio for a given change in ionization rate.

(c) Calculations of track diameters based on the observed etching rate versus ionization relation for an ordinary glass agree well with recent measurements.

(d) The etching rate V_T along tracks in crystalline inorganic solids is undoubtedly also an increasing function of ionization rate, although the function will probably depend on crystallographic direction as well as on ionization rate.

ACKNOWLEDGMENT

This work was supported by the National Aeronautic and Space Administration under Contract No. NAS 9-7898.

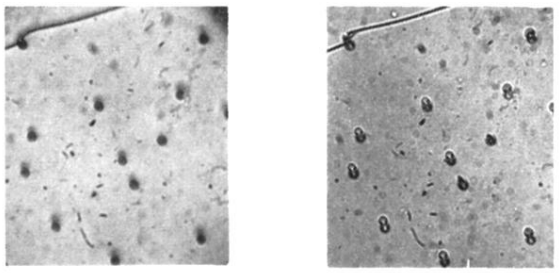

FIG. 7. Decrease in etching rate with slowing down of Cf^{252} fission fragments. Left: etched tracks on upper surface of thin sheets of SiO_2 glass viewed in reflected light. Right: the same etched tracks on lower surface viewed in transmitted light. Decreases in ionization rate during slowing down are revealed by smaller diameters of the pits on the lower surface.