Formation of Etchable Tracks in Dielectrics*

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It is proposed that etchable damage is produced in dielectrics by energetic charged particles when a critical dosage of ionization energy is deposited at a critical distance from the ion's path by secondary electrons. Within the critical cylinder, molecular fragments more soluble than the parent molecule are formed. The radius of the critical cylinder is taken to be approximately 20 Å, as is appropriate to the passage of the etchant along the track and the diffusion of reaction products back to the surface. At the critical radius the dosage approximates doses producing bulk damage under γ irradiation. The proposed criterion predicts the formation or nonformation of etchable tracks in Lexan polycarbonate, cellulose nitrate, and mica, in agreement with published data. The calculations have been extended to magnetic monopoles to establish criteria for their detection in dielectric track detectors.

I. INTRODUCTION

SINCE the observation of fission fragment tracks in mica by Silk and Barnes[1] in 1959, by use of an electron microscope, a number of investigators have studied the process of track formation in dielectrics, and a variety of mechanisms (thermal spike, displacement spike, ion-explosion spike) have been suggested for their formation. The applicability of these detectors to physical problems is enhanced by their relative insensitivity to electrons and γ rays, and by the fact that the tracks of heavy ions are readily etched to optical dimensions. Many of these points are discussed in a 1965 review article by Fleischer, Price, and Walker.[2]

Precise calibration of these detectors with heavy-ion beams is possible with available accelerators (HILAC) only up to argon at 10 MeV/amu. To extrapolate to ions of higher mass and energy, significant for the detection of the heavy primary cosmic rays, several correlative track formation criteria have been proposed, including the rate of energy loss of the bombarding ion (appropriate to a thermal-spike criterion),[3] the rate of primary ionization (appropriate to an ion-explosion spike criterion),[4] and the rate of restricted energy loss (energy loss to electrons acquiring less than 1 keV of kinetic energy).[5]

II. DISCUSSION

According to the present view, the mechanisms of track formation differ according to the ion energy. Near the end of the range of a heavy ion, at energies below approximately 0.05 MeV/amu, the principal mode of energy loss is by nuclear collision. Here, the displacement spike model is appropriate, as in the case of α-recoil tracks observed by Huang and Walker.[6] At higher ion energies the principal mode of energy loss is through ionization, and the track is formed through the deposition of the energy of secondary electrons (δ rays) in the immediate vicinity of the ion's path. Molecular fragments resulting from interaction of δ rays with the surrounding medium are more soluble than the parent molecules. An etchable track is formed if sufficient fragmentation has occurred over a sufficiently large diameter so that the etchant can pass freely down the damaged trail into the body of the dielectric.

These considerations lead to a criterion of minimal dosage of deposited ionization energy at a minimal diameter.

In the tangle of δ rays in the immediate vicinity of the ion's path, the production of ionizing events may be approximated as random and the response of the medium may be represented by the Poisson distribution. The hypothesis that many fragments must be generated from a large parent molecule by bond-rupturing ionizations characterizes the process as a many-hit event. Since the cumulative Poisson distribution approaches a step function as the required number of hits increases, the required dosage is taken to have a threshold or step-function character. Thus a critical dose is required for the production of an etchable track. The spatial distribution of ionization energy about the path of a bombarding ion must first be determined. These calculations are described in a preceding paper by the present authors.[7]

The critical diameter for track formation is limited from below by the size of the solvent complex, which may be estimated as approximately 5 Å in diam. The critical diameter is limited from above by experimental observation of the size of the damaged region. Electron

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[1] E. C. H. Silk and R. S. Barnes, Phil. Mag. **4**, 970 (1959).

[2] R. L. Fleischer, P. B. Price, and R. M. Walker, Ann. Rev. Nucl. Sci. **15**, 1 (1965).

[3] R. L. Fleischer, P. B. Price, R. M. Walker, and E. L. Hubbard, Phys. Rev. **133**, A1443 (1964).

[4] R. L. Fleischer, P. B. Price, and R. M. Walker, J. Appl. Phys. **36**, 3645 (1965).

[5] E. V. Benton, U.S. Naval Radiological Defense Laboratory, Report USNRDL-TR-67-80, 1967 (unpublished).

[6] W. H. Huang and R. M. Walker, Science **155**, 1103 (1967).

[7] E. J. Kobetich and R. Katz, second preceding paper, Phys. Rev. **170**, 391 (1968).

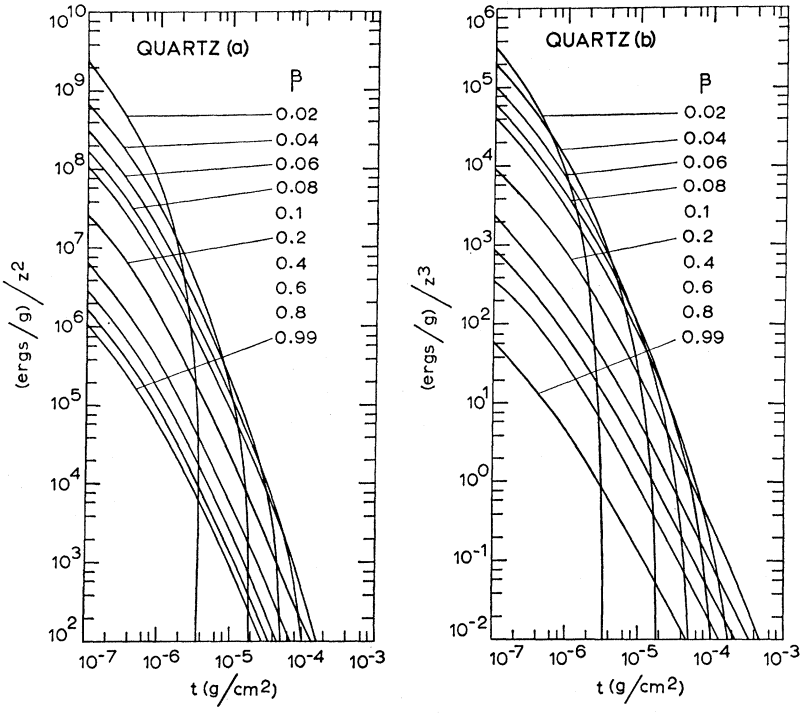

FIG. 1. Spatial distribution of ionization energy in quartz. To find the energy dosage deposited at radius t (erg/g) by an ion of effective charge number z moving at speed βc, the value given in curve (b) must be multiplied by z, added to the value obtained from (a), and the sum multiplied by z^2. The effective charge number may be obtained from the atomic number Z and speed by the expression $z=Z[1-\exp(-125\beta Z^{-2/3})]$. See Ref. 7.

microscope observations of Silk and Barnes[1] of fission fragment damage in mica set an upper limit of about 100 Å, while similar observations by Chadderton and Montagu-Pollock[8] give a value of about 150 Å for fission fragment damage in other crystals. Chemical etching experiments of Price and Walker[9] imply a track diameter not greater than 50 Å in mica. In the present work dosage of deposited ionization energy at 20 Å from the ion's path has been used to characterize the process. Since no data exist on the energy dissipa-

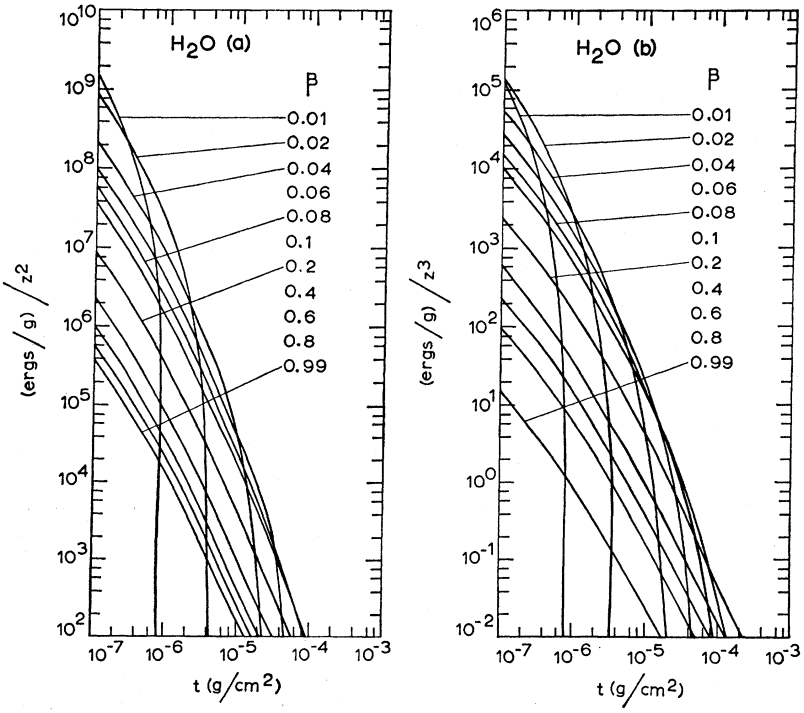

FIG. 2. Spatial distribution of ionization energy in water. See caption for Fig. 1.

[8] L. T. Chadderton and H. M. Montagu-Pollock, Proc. Roy. Soc. (London) **A274**, 239 (1963).
[9] P. B. Price and R. M. Walker, J. Appl. Phys. **33**, 3407 (1962)

tion of electron beams in films of 20 Å thickness, the formulation of the dissipation of the energy of electron beams shown in a preceding paper[7] to be valid from 1 keV to 2 MeV is extrapolated to electrons of energy down to 20 eV for these needs.

Further, it must be anticipated that the critical dosage is comparable to the γ-ray dose capable of producing macroscopic damage in the bulk material, for the mechanism of damage production in both cases is through the interaction of secondary electrons with the medium. A dosage capable of producing extensive molecular fragmentation for etchable damage must also result in marked deterioration of the physical properties of the bulk material.

A large fraction of the energy deposited at distances of 20 Å from the ion's path arises from δ rays whose energy lies below 0.1 keV. From the δ-ray distribution formula,[7] it is easy to see that the fraction of primary ionization lying in this energy region remains substantially constant at ion energies above a few MeV/amu; the precise limit depending on the mean ionization potential of the medium. Thus the correlative criteria for track formation, of total primary ionization suggested by Fleischer, Hubbard, Price, and Walker,[10] and of restricted energy loss rate suggested by Benton[5] are consistent with the present theory.

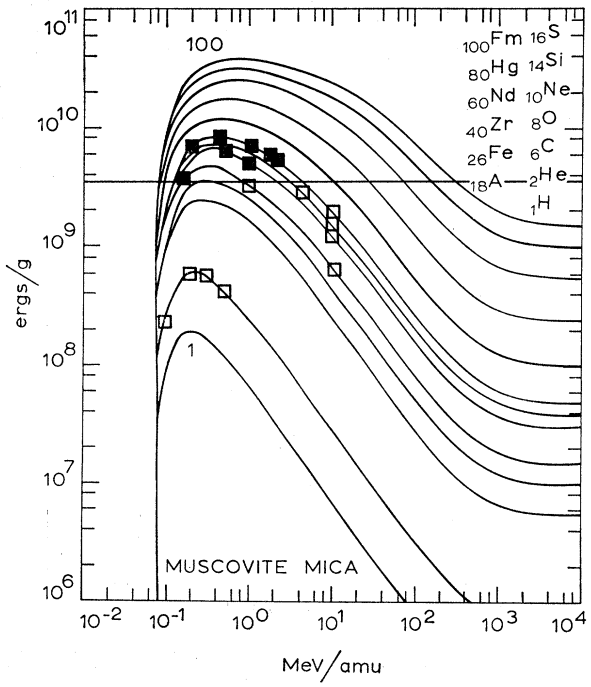

FIG. 3. Dosage of ionization energy in mica at 19 Å (quartz at 20 Å) as a function of bombarding ion and ion energy. Published data (see Ref. 10) are plotted as solid circles for bombardments at which etchable tracks are formed, and as open circles otherwise. Bombardments where partial track formation has been observed lie along the dividing line of critical dose and have not been plotted.

[10] R. L. Fleischer, E. L. Hubbard, P. B. Price, and R. M. Walker, Phys. Rev. **156**, 353 (1967).

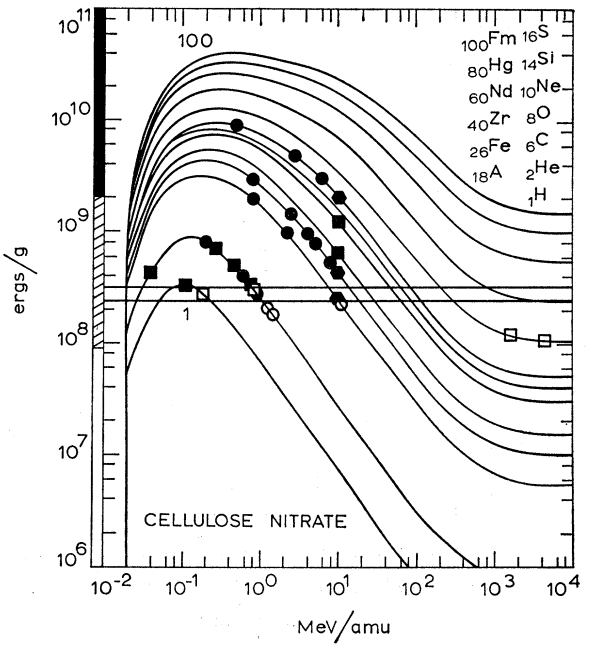

FIG. 4. Dosage of ionization energy in cellulose nitrate at 15 Å (water at 20 Å). Published data from Fleischer *et al.* (see Ref. 10) are plotted as solid squares if etchable tracks are formed and as open squares if not. Data from Benton (see Ref. 5) are similarly plotted as circles. Overlapped points are plotted as hexagons. Shading along the abscissa gives the response of bulk cellulose nitrate to γ irradiation. An open bar implies that the dosage yields negligible damage, while cross hatching implies moderate damage and the solid bar implies severe damage. Adjacent thresholds for the two sets of data may imply manufacturing variations in the plastic material.

A simple calculation of the electrostatic energy stored in the ionized core, after the passage of a heavy ion through a dielectric medium, shows that the energy per atom within the core is substantially less than the energy needed to produce a Frenkel defect; even at 10 Å from the ion's path in Lexan polycarbonate.This suggests that the ion-explosion spike mechanism for track formation is invalid, though the criterion of total primary ionization initially motivated by this mechanism correlates well with available data.

III. RESULTS

Within variations which may be neglected for the present purposes, the atomic composition and therefore the mean ionization potential of silicate minerals is nearly the same. For this reason the spatial distribution of ionization energy in quartz, shown in Fig. 1, is used to represent all silicate minerals. Similarly, the spatial distribution of ionization energy in water, shown in Fig. 2, is used to represent all plastics. The dose of deposited ionization energy in quartz at 20 Å from the path of an ion of atomic number and energy indicated on the figure is shown in Fig. 3. Similar results for water are shown in Figs. 4 and 5. Because of density differences Fig. 3 corresponds to energy deposi-

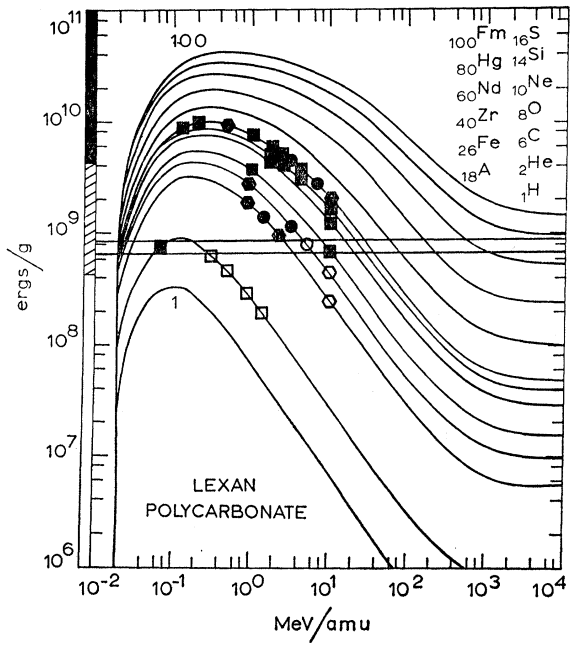

FIG. 5. Dosage of ionization energy in Lexan polycarbonate at 17 Å (water at 20 Å). Published data from Fleischer *et al.* (see Ref. 10) are plotted as solid squares if etchable tracks are formed and as open squares if not. Data from Benton (see Ref. 5) are similarly plotted as circles. Overlapped points are plotted as hexagons. Shading along the abscissa gives the response of bulk polycarbonate to γ irradiation, as in Fig. 4.

tion in mica at 19 Å, Fig. 4 to cellulose nitrate at 15 Å, and Fig. 5 to Lexan polycarbonate at 17 Å.

Data reported by Fleischer *et al.*[10] on the formation of etchable tracks in mica are plotted on Fig. 3. Solid squares represent bombardments at which etchable tracks are observed, while open squares represent bombardments at which no tracks are observed. A horizontal line at an energy dosage of about 3.5×10^9 erg/g at 19 Å from the ion's path in muscovite mica clearly separates the region of track formation from the region of nonformation. The calculation predicts that no relativistic ion will form etchable tracks in mica. The length of tracks in mica sheets may be inferred from the energies at which the energy deposition curves cross the critical dosage line. It is interesting to note that a $_{40}$Zr fission fragment at an energy of 1 MeV/amu has an effective charge of 15.5 and produces the critical dose at $\sim$60 Å from its path, from Fig. 1, in agreement with electron-microscope observation of the extent of the damaged region.

Similar results are obtained for cellulose nitrate and Lexan polycarbonate. Data from Fleischer *et al.*[10] are plotted as squares, data from Benton[5] are plotted as circles, while overlapped points are plotted as hexagons in Figs. 4 and 5. Again, solid figures represent the observation of etchable bombardments, while hollow figures represent bombardments at which no etchable tracks are formed. In these figures pairs of lines are drawn, one separating solid squares from hollow squares, and the other separating solid circles from hollow circles. All data were obtained with accelerator bombardments except two hollow squares on Fig. 4, representing the failure to detect relativistic iron nuclei in the cosmic rays with cellulose-nitrate detectors.

In Figs. 4 and 5, bars have been drawn along the ordinate to represent the γ ray dose for macroscopic damage of the bulk materials, as reported by the Radiation Effects Center of the Battelle Memorial Institute.[11] The boundary between the open and cross-hatched bars is the average dose for threshold damage, while the boundary between cross-hatched and solid bars represents a reduction of 25% in a collection of physical properties. Doses covered by the open bar then represent negligible damage; the cross-hatched bar represents moderate damage, while the solid bar represents the region of severe damage. For both cellulose nitrate and Lexan polycarbonate the critical dosage for etchable track formation lies in the region of moderate damage. No comparable data are available for mica.

The presence on these two curves of two different but adjacent thresholds, for what is nominally the same material, may reflect variations in the manufacturing process, and implies that each batch of plastic must be separately calibrated.

It is pertinent to ask to what extent is the 20 Å criterion a firm one. Clearly, some relaxation in this number is possible. But if the energy-deposition calculation is assumed to be correct, the critical diameter cannot be relaxed very much. The dosage falls off to zero very rapidly at low ion energies in Figs. 3–5. This arises from the range of the δ ray of maximum energy at the

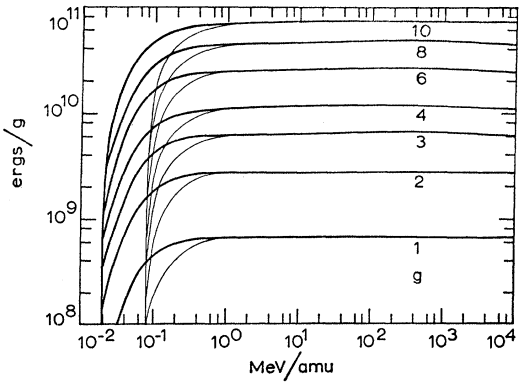

FIG. 6. Dosage at 20 Å in quartz (light lines) and in water (heavy lines) produced by magnetic monopoles of pole strength $g=n(137e/2)$. Comparison with Fig. 4 indicates that cellulose nitrate will yield etchable tracks if bombarded with relativistic Dirac monopoles ($n=1$).

[11] R. W. King, N. J. Broadway, and S. Palinchak, REIC Report No. 21 (1961), Radiation Effects Information Center, Battelle Memorial Institute, Columbus, Ohio 43201 (unpublished).

indicated ion energy. No energy can be deposited beyond the range of the δ ray of greatest energy. Thus, plotted profiles of the energy dosage at 25 Å (not shown) yield discrepant points at low ion energies. The critical diameter of 40 Å used in the present work is based upon internal consistency as well as upon external considerations mentioned earlier.

In view of the continuing interest in the possible detection of magnetic monopoles in cosmic rays, the dose of ionization energy at 20 Å from the path of a magnetic monopole has been calculated for water and for quartz for a selection of magnetic charges shown in units of the pole strength of the Dirac monopole, $g=n(137e/2)$, as shown in Fig. 6. These calculations imply that a Dirac monopole of unit strength is detectable in cellulose nitrate, even at relativistic energies.

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Width of Heavy-Ion Tracks in Emulsion*

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Profiles of the solid core of long, flat, ending tracks of heavy primary cosmic rays in Ilford G-5 emulsion have been measured by manually tracing their enlarged (3500×) photographic image. These measurements agree well in the ending 3000 μ of residual range ($\beta<0.3$) with a reformulated theory of track width based on computation of the spatial distribution of ionization energy. The measured core width in G-5 emulsion is the sum of the sensitized cylinder diameter, at which 6000 erg/cm^3 of ionization energy is deposited by δ rays, and the diameter of a developed grain.

I. INTRODUCTION

In 1965, Katz and Butts[1] showed that the distribution of ionization energy, about the path of an energetic ion, provided a suitable basis for analyzing the core width of its track in electron sensitive emulsion. In the intervening time, a number of the components of the calculation have been improved substantially. The dissipation of the energy of normally incident electrons in plane films has been reformulated empirically[2] so as to eliminate the original assumptions of a power-law range-energy relation, and of straight-line electron paths. The δ-ray distribution formula has been amended to include atomic binding effects. Similar progress has been made in track measurement through use of photomicrographic techniques. The result has been a considerable improvement in the agreement between theory and experiment, to the point that the theory describes measured tracks well, to a residual range of about 3000 μ ($\beta<0.3$). The principal conclusion of the earlier work, that the track core is defined by the cylinder diameter at which the deposited ionization energy is 6000 erg/cm^3, in Ilford G-5 emulsion, remains unchanged. The theory approximates the emulsion as a homogeneous medium, treats the photographic grain as a point particle, and employs the concept of an energy-deposition threshold for the sensitization of a photographic grain. The validity of these approximations improves as the ratio of the diameter of the sensitized cylinder to the grain diameter increases, so that the theory provides a basis for the identification of ending tracks of very heavy primary cosmic rays in G-5 emulsion.

II. TRACK MEASUREMENT AND CALIBRATION

Two stacks of Ilford G-5 emulsion flown at balloon altitudes from Ft. Churchill at different times, and processed in different laboratories to approximately the same grain diameter (0.7 μ) were scanned for long, flat, wide, ending tracks with a Bausch and Lomb Stereo-Zoom microscope at 40×. Track segments 50 μ long were photographed, under photometric control, onto 100-ft rolls of 70-mm Kodak LS Pan film with a Leitz microscope equipped with a xenon lamp and a 100× Leitz Plano objective. Film rolls were processed and enlarged commercially onto 8-in.-wide rolls of Kodak Velox Premier paper, under photometric control, to a resultant enlargement of 3500×.

* Supported by the U.S. Atomic Energy Commission and the National Science Foundation.

[1] R. Katz and J. J. Butts, Phys. Rev. **137**, B198 (1965).

[2] E. J. Kobetich and R. Katz, third preceding paper, Phys. Rev. **170**, 391 (1968).