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# Particle Track Etching

Diverse technological uses range from virus identification to uranium exploration.

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In this article we describe a case in which the study of basic science has led to practical results—the case of particle track etching (1–4). We believe that the extreme versatility of this technique points out the essential unpredictability of the consequences of science and hence the opportunities inherent in understanding how things work.

In particular, we will describe here how the simple discovery of track etching has led to applications in semiconductor electronics, aerosol sampling, the identification of microbiological par-

ticles, nuclear engineering, mine safety, neutron radiography, uranium exploration, tracer technology, the determination of the color of gem diamonds, the clarification and stabilization of beer and wine, and a variety of peaceful uses for nuclear reactors. This list is but a sampling of a larger list, intended to convey the diversity of possibilities.

## Observation of Particle Tracks

As a charged particle such as a fission fragment moves through a solid, it produces a region of permanent, catastrophic atomic damage (5). Examples of such damage tracks, first seen directly in transmission electron microscopy (6), are shown in Fig. 1A. The critical step that made it possible to use these tracks was the discovery that the damage could be preferentially dis-

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solved chemically and the resultant hole enlarged to optical size. This technique was first demonstrated on micas (7) and subsequently on glass (8) and plastics (9), so that now an extensive library of materials and etchants exists (10). Figure 1, B to D, shows tracks in the three major classes of solid materials (8, 9, 11).

These tracks are useful primarily for three reasons: (i) the technique is

simple; (ii) a geometrical shape (a hole) results; and (iii) a sharp threshold exists between particles that leave tracks and those that do not (12). Because of item (iii), it is possible for the investigator to ignore a high background of uninteresting particles; as we shall note later, item (ii) leads to a method of particle identification (13).

We now summarize the technological applications of etched particle

tracks—the term “application” is defined here as a use that is of potential economic or public good. The scientific aspects of etched particle tracks—such work has resulted in several hundred publications in such diverse fields as nuclear physics, geophysics, geochronology, archeology, lunar and meteoritic physics, and cosmic-ray physics—are deliberately excluded (1–4, 13). We divide applications into four categories: (i) porous membranes, that is, filters and particulate counters; (ii) imaging and mapping; (iii) radiation dosimetry; and (iv) miscellaneous.

### Porous Membranes

In one of its simplest applications, particle track etching is used to perforate thin sheets of material to produce filters with nicely geometrical holes and closely controlled porosity and hole size (14, 15). A thin detector material is exposed to a collimated beam of particles that produces tracks across the entire thickness of the sheet. The subsequent etching produces holes whose diameters can be controlled by specifying the etching time, and whose numbers are determined by the chosen particle dose. Hole diameters ranging from 50 angstroms (7, 16) on up (14) can be made in mica and various plastic materials. In the commercial embodiment (called Nuclepore), polycarbonate film is irradiated with  $^{235}\text{U}$  fission fragments in a nuclear reactor and later etched in a warm sodium hydroxide solution to give holes such as are shown in Fig. 2B. By comparison, the conventional fibrous filter shown in Fig. 2A does not have the simple, sharp geometry that is desirable in many applications.

Etched filters have been put to diverse uses. These etched filters [see (2), cover photograph, or Fig. 11] were first used to separate free-floating cancer cells from blood (17), making use of the fact that cancer cells are both larger and more rigid than normal blood cells. Seal demonstrated (17) that malignant cells in the blood could be separated and recognized; it has not yet been established whether such cells become present in the blood early enough in the development of the cancer that their source can be identified and treated effectively.

In other cytological applications the well-defined hole sizes of track-etched filters have been used to isolate and

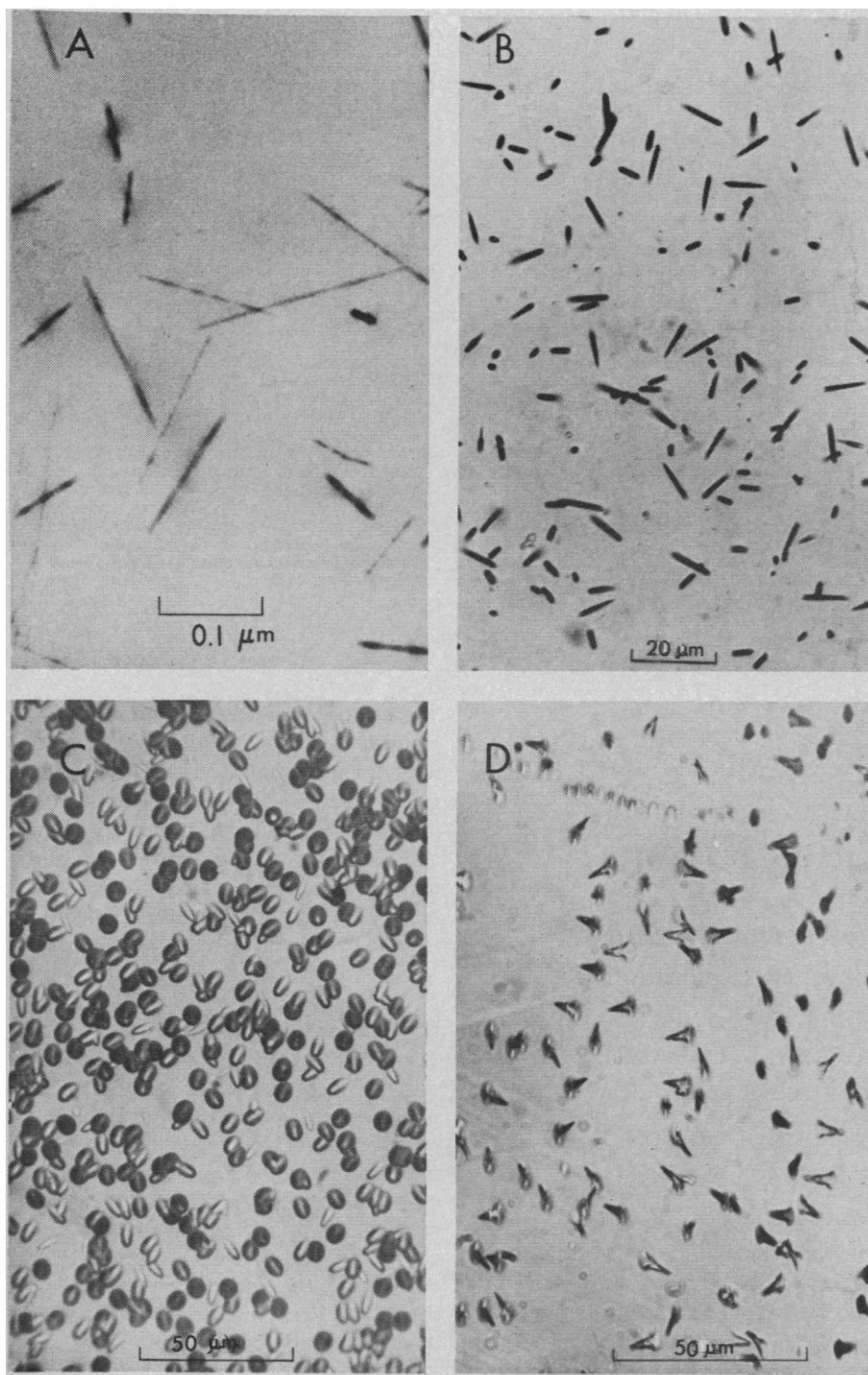


Fig. 1. Fission fragment tracks in (A) mica, seen directly with an electron microscope (70); etched particle tracks viewed optically in (B) a polymer (9), (C) a glass (8), and (D) a crystal (11).

examine cells of known size and measure their deformability (18). Nuclepore filters have also been found to be effective devices for aerosol filtration in environmental protection applications (19), for cleaning gases used for dusting electronic components prior to encapsulation (20), and for producing ultrapure solutions for use in the fabrication of semiconductor devices.

Nuclepore (20) has been used to clarify and stabilize wine and beer (by removing bacteria). Such stabilization makes it possible for draft beer to be stored safely at room temperature, with no need for the heating that otherwise would be required to destroy the bacteria in bottled beer, a heating that is responsible for the differences in taste between draft beer and the conventional bottled variety.

One of the most ingenious and economical uses of etched tracks is the DeBlois-Bean counter (21) in which a single etched track is used to count and size small particles in an electrolyte. This counter extends the principle and uses of the Coulter counter (22). As shown in Fig. 3A, the two sides of an electrochemical cell are separated by a membrane with a track etched through it. The resistance between the electrodes depends primarily on the conducting path through the hole. As a charged insulating particle enters, the resistance increases in proportion to the volume of the particle. At the same time the velocity of the particle through the hole is a measure of its charge, so that these two quantities—size and charge (or mobility)—can be used to characterize and identify individual particles. The oscilloscope traces in Fig. 3B show, for example, how clearly different are the signals of polystyrene spheres 910 angstroms in diameter from those of T2 viruses with an effective average diameter of 1010 angstroms. It is expected that the equipment available at present will make sizes down to 300 angstroms accessible, with automatic readout of the results.

A bibliography on filtration with the use of etched particle tracks is available from the authors (23).

### Imaging and Mapping

Track detectors have such a variety of sensitivities that some of them can record nuclear particles ranging from fission fragments down to alpha particles (2, 12), deuterons (2), and protons

(24). However, they are *not* sensitive to lightly ionizing radiation. These characteristics have led to new types of radiographic applications. In some cases the tracks are measured individually; in

other cases where enough tracks are present, they are used to give a visual image much like a photograph. It is also possible to map neutrons by recording alpha particles or fission frag-

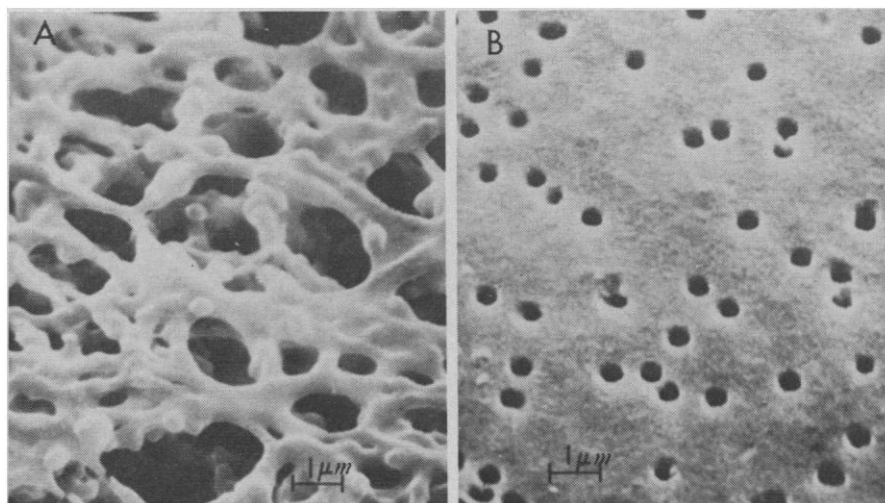


Fig. 2. Scanning electron micrographs of fine hole size filters: (A) cellulose fiber filter; (B) filter produced by particle track etching. Effective size for the fiber filter, 0.45 micrometer; hole size for the etched filter, 0.4 micrometer.

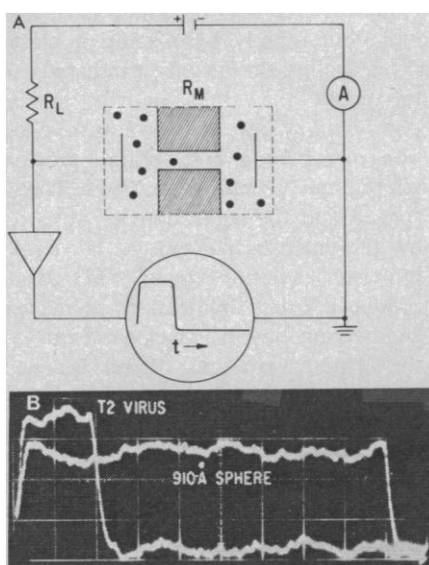
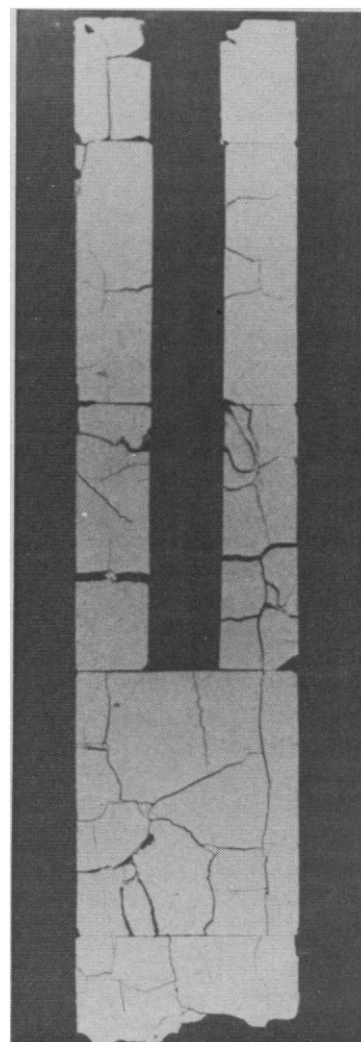


Fig. 3 (above). (A) Sketch of the DeBlois-Bean counter: a membrane with a single etched hole is immersed in an electrolyte with electrodes on either side;  $R_L$ , external resistance;  $R_M$ , internal resistance;  $t$ , time;  $A$ , ammeter. (B) Oscilloscope traces as polystyrene sphere and T2 virus transit the hole. Individual particles are distinguishable and are identified by their size (given by the pulse height) and charge (inferred from the transit time) (21). Fig. 4 (right). Alpha radiograph of a mixed uranium oxide-plutonium oxide fuel rod shows cracking produced by thermal stresses. The image consists of etched tracks of alpha particles from plutonium and, to a lesser extent, uranium; a gamma- or beta-radiograph would be fogged.



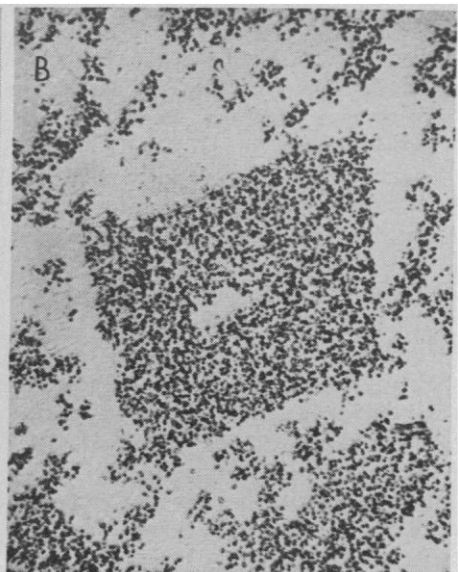
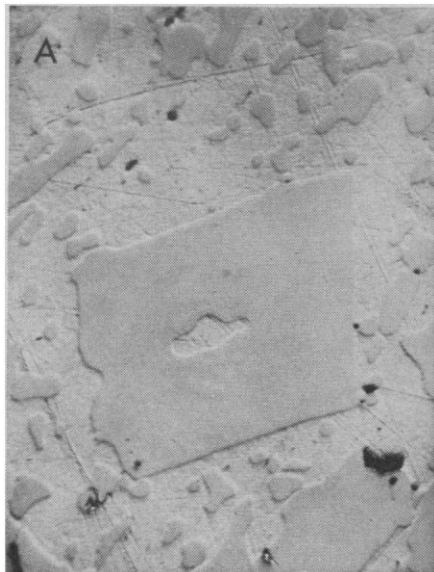
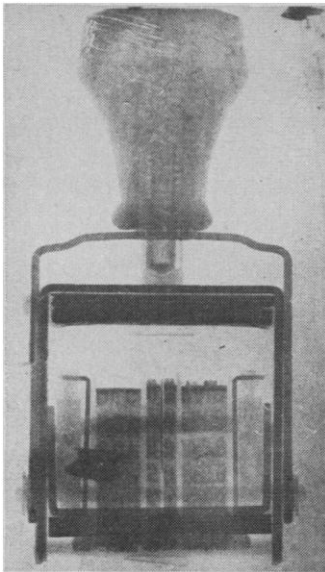


Fig. 5 (left). Neutron radiograph of a rubber date stamper recorded by alpha-particle tracks in a cellulose nitrate sheet with an adjacent boron activation plate that supplies alpha particles from  $(n, \alpha)$  reactions. [Courtesy of C. R. Porter, General Electric Vallecitos Nuclear Center] Fig. 6 (right). Boron-rich particles in stainless steel: (A) polished etched surface of stainless steel; (B) alpha-particle map of the same area showing the boron-rich sites as dark regions consisting of light-scattering, etched alpha-particle tracks from  $(n, \alpha)$  reactions. The large particle (shaped like a parallelogram) is 75 micrometers wide (31). [Courtesy of North-Holland Publishing Company, Amsterdam]

ments from neutron-induced reactions such as  $^{10}\text{B}(n, \alpha)^7\text{Li}$  or  $^{235}\text{U}(n, \text{fission})$ . Applications extend from nuclear engineering to superconductivity, metallurgy, mining exploration, and tracer studies.



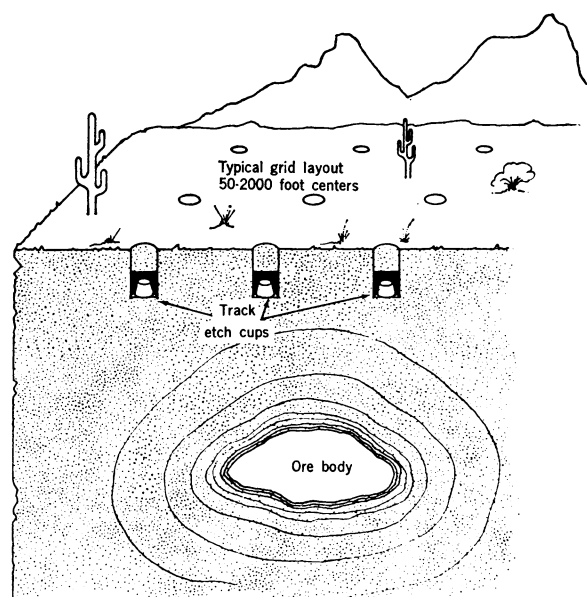
Fig. 7. Micrograph of the uranium distribution in taper-sectioned wires (0.005 centimeter wide) with uranium-enriched tin diffused into a thick outer section of the niobium wire to produce a layer of  $\text{Nb}_3\text{Sn}$ . Induced uranium fission tracks clearly delineate the thickness of the uranium-rich layer, which is the same as that of the  $\text{Nb}_3\text{Sn}$  sheet.

In one of the earliest examples of alpha radiography the distribution of  $^{239}\text{Pu}$  in a uranium oxide-plutonium oxide fuel rod was measured (25). This was done simply by placing a plastic sheet against the heavily irradiated rod to record the alpha activity of the plutonium. Figure 4 shows an application in which a conventional nuclear photograph would have been heavily fogged and the alpha activity obscured by the intense background of lightly ionizing radiation from the fission products. Other applications in nuclear engineering involve measurements of the spatial variations of reactor neu-

tron fluxes (26) and reaction rates in reactors (27).

Track-based neutron radiography can be performed with a detector assembly consisting of a boron-rich plate pressed against a plastic detector sensitive to alpha radiation such as cellulose nitrate or one of the cellulose acetates (28). A beam of thermal neutrons is passed through the material to be radiographed and is then imaged by the alpha-particle tracks resulting from the  $^{10}\text{B}(n, \alpha)$  reaction. When etched, these tracks are light-scatterers, so that the plastic sheet itself gives a direct, visual image of the neutron transmission

Fig. 8. Uranium exploration technique. Radon-222 moving upward from the decays of uranium deposits by alpha emission in buried, inverted cups, leaving tracks in cellulose nitrate detectors. In a typical grid layout there is a 50-foot (15.5-meter) separation between the center of one cup and its nearest neighbor. The contours of the track density reveal likely sites for more detailed mining exploration (35).



through the object. Neutron radiography is a complementary technique to normal x-radiography, being in general more sensitive to lighter elements, in particular, hydrogen, and hence is specially useful where organic material is to be examined, whether it be explosives or biological tissue. Track etch plates are uniquely useful in high radiation environments, where other types of plates are readily fogged. They have the additional special virtue of high resolution, since the limiting detail available is set by track lengths, typically a few micrometers for neutron radiographic applications. Radiographs can be taken with as few as  $2 \times 10^7$  neutrons per square centimeter and can reveal thickness differences of 1 percent (29). Figure 5 gives an example of a neutron radiograph of a rubber date stamper. Strong neutron scattering by organic materials such as the wooden handle and the rubber belt provide good contrast for materials that are rich in hydrogen.

The imaging properties also provide a powerful tool for the recognition of "tagged" bits of particulate matter. Even a submicroscopic particle with a modest concentration of uranium, boron, or lithium will produce a characteristic track "star" when it is irradiated with neutrons (11, 30). Thus the motion of particles released in one location can be followed over long distances even after the particles have suffered great dilution. Particles can be given distinctive characters if, for example, their ratio of boron to uranium is specified and if the resulting ratio of alpha tracks to fission tracks is used to characterize the composition of the recovered particles.

The chemical mapping of boron, lithium, nitrogen, and oxygen by induced  $(n, \alpha)$  or  $(n, p)$  reactions or of heavy elements such as uranium, plutonium, and thorium by induced fission reactions is a powerful metallurgical tool (31-34). The dominant element in stainless steel that emits charged particles during neutron bombardment is boron; its distribution can be readily mapped on an alpha-sensitive detector, as displayed in Fig. 6 (31). The same technique has been used by Chrenko to demonstrate that the color of gem diamonds is correlated with the boron content (32).

Basically the same technique was used in a study of the high-field superconducting properties of intermetallic compounds containing boron or uranium additions to be used to produce

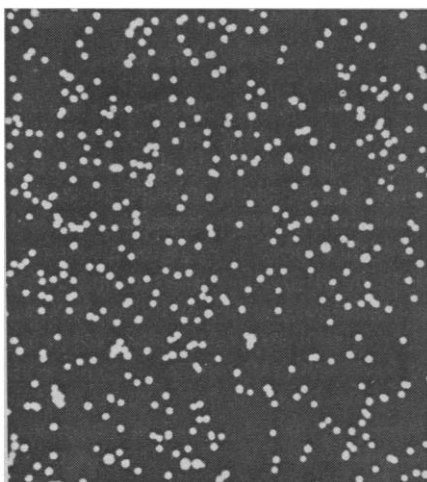


Fig. 9. Track contrast enhancement. Track-etched holes in a special red film show high contrast when viewed with green light (44).

flux-pinning sites (irregularities that hinder the motion of magnetic field lines) as a result of neutron reactions. These reactions allowed the state of dispersion of the boron and uranium to be visualized. In one case (33) it was shown that the boron was not dispersed on an atomic scale and hence would not be effective. In contrast, the doping of a  $\text{Nb}_3\text{Sn}$  coating on niobium wires was shown to have been successful on the basis of the micrograph in Fig. 7, which shows that uranium has diffused to a depth that is equal to the thickness of the outer sheath of  $\text{Nb}_3\text{Sn}$ .

A particle track technique is being employed in field exploration for uranium ores (35). One member of the  $^{238}\text{U}$  decay chain,  $^{222}\text{Rn}$ , is a gas, with therefore the possibility of rapid motion through permeable geological deposits

and higher surface emanation over uranium-rich deposits. The detection method, outlined in Fig. 8, is to use buried track detectors to register alpha particles from the radon decay. By statistical analysis of the observed track densities, contour maps may be constructed to show the regions of high radon emanation and hence to identify sites for possible exploratory drilling.

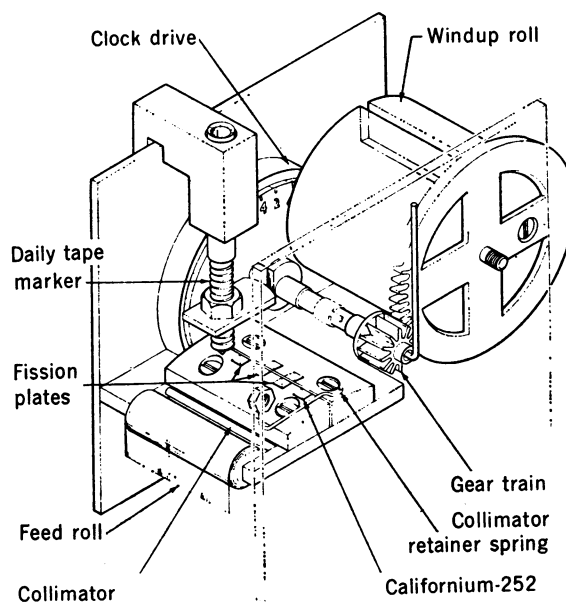
A bibliography on reactor physics and particle imaging applications of particle track etching is available from the authors (23).

### Etched Particle Track Enhancement and Counting

Doses of many charged particles can be measured directly by the use of track detectors (12), and various other particles may be measured indirectly if induced reactions are used to produce charged particles that may be recorded (26, 36). Thus low-energy ( $\approx 5$  million electron volts) alpha particles can be seen, and ions of atomic number  $\approx 30$  can be observed even at relativistic energies (37). Low-energy heavy particles, such as fission fragments, are readily recorded (7).

Indirect dosimetry methods most commonly record alpha particles or fission fragments. For example, such particles are released by  $(n, \alpha)$  or  $(n, \text{fission})$  reactions caused by thermal neutrons;  $(n, 3\alpha)$  reactions provide distinctive three-pronged events that can be used to measure fast neutron fluences (3), and  $(n, \text{fission})$  reactions on elements that do not fission after exposure to thermal neutrons can be

Fig. 10. Nuclear safeguards device to monitor use of nuclear reactors. The neutron energy spectrum is recorded as a function of time by fission tracks induced in  $^{235}\text{U}$ ,  $^{238}\text{U}$ ,  $^{237}\text{Np}$ , and  $^{232}\text{Th}$  and recorded on a moving polycarbonate tape. A  $^{252}\text{Cf}$  fission source provides a known flux of particles to permit calibration of the tape speed (48).



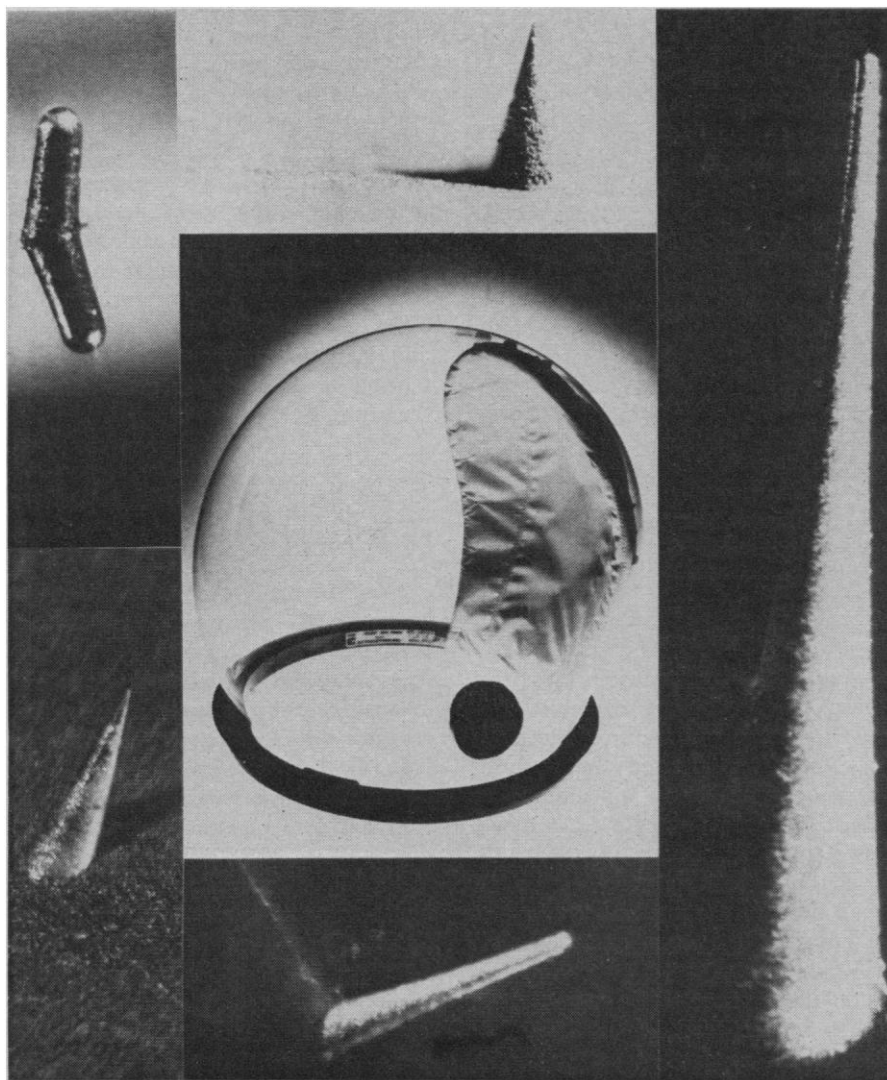
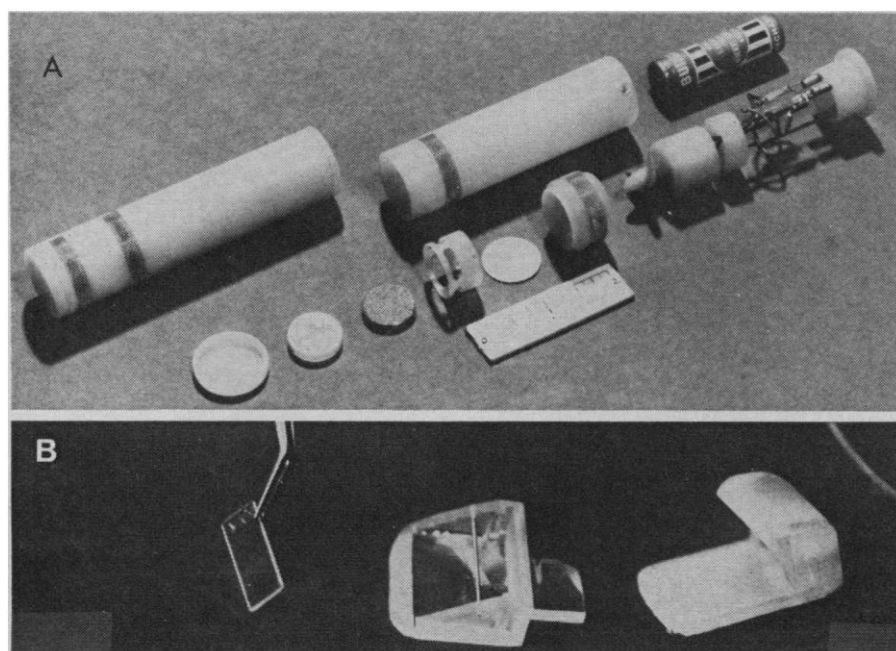


Fig. 11. Apollo helmet and cosmic-ray particle track replicas. The helmet was worn by astronaut James Lovell in the Apollo 8 mission; the tracks were caused by heavy cosmic rays that penetrated the Apollo helmets and provide a measure of the cytologically lethal component of the cosmic radiation (49).



used to measure doses of fast neutrons (38), high-energy charged particles (3, 39), or photons (40).

Although the prime dosimetry information is displayed by etched particle tracks, readout which involves counting individual tracks through a microscope can be a laborious process; consequently dosimetry applications have taken on new impetus with the development of simple, automatic methods of track counting. In the most elegant of these, a d-c voltage is applied to a sandwich of etched track material and a thin aluminum electrode (41). As the voltage increases, sparks occur in the individual tracks. The energy in the spark is sufficient to vaporize the aluminum electrode in the vicinity of the track hole, and, as a result, each track gives just one spark. The total number of sparks can be recorded by means of a simple scaler. Alternatively one can use a light meter to measure the optical transmission through such foils; this procedure provides a quick method for the rapid intercomparison of samples. In a recent development of this technique, nanosecond pulses were used to give reliable spark counts of thin samples (42). At low doses, where track densities are correspondingly low, a number of additional methods of locating and counting a few tracks etched through large detector areas have been developed. In the quickest and simplest of these a conventional Ozalid machine is used: ammonia gas passes through the holes and stains an ammonia-sensitive paper, so that, by counting large blue dots on a piece of paper, one can make rapid track density measurements (43). A special detector film has been devised by Kodak to facilitate rapid readout (44). In this material a red layer (8 micrometers thick) of intensely dyed cellulose nitrate is laminated to a clear backing material. The etching of tracks through the red layer removes the coloring. When viewed through an appropriate green filter, light is transmitted only where the presence of a track has allowed the dye to be removed. The

Fig. 12. Two alpha-particle dosimeters for use by uranium miners: (A) an active device that strains aerosol particles from mine air to test for the alpha radioactivity of  $^{222}\text{Rn}$  daughter products (54) (the scale is in inches); (B) a passive device that records the alpha activity of both  $^{222}\text{Rn}$  and its daughters (55). [Fig. 12A courtesy of Pergamon Press, Oxford, England]

result, as seen in Fig. 9, is a high-contrast "starfield" of lighted tracks on a black background, suitable for various rapid readout methods.

A bibliography on etched particle track enhancement and counting is available from the authors (23).

### Specific Dosimetry Applications

Neutron dosimetry is perhaps the most extensively utilized type of track dosimetry. Neutrons can be measured either by the fissions they induce in, for example,  $^{232}\text{Th}$ ,  $^{235}\text{U}$ , or  $^{237}\text{Np}$  or by  $(n, \alpha)$  reactions of (usually)  $^{10}\text{B}$ . Both  $^{235}\text{U}$  and  $^{10}\text{B}$  are sensitive to thermal neutrons. By using uranium-loaded glasses in conjunction with different types of track detectors (26), investigators in laboratories around the world are now routinely monitoring thermal neutron doses. Standard uranium glasses have been fabricated for this purpose by both Corning Glass Works (45) and the National Bureau of Standards (46).

Personnel dosimetry devices of several types have also been fabricated. In sophisticated versions, various targets including  $^{232}\text{Th}$  and  $^{237}\text{Np}$  (only fast neutrons can be used to bring about the fission of these isotopes) are used to give a measure of the neutron energy spectrum (38). Detectors of this type now form part of the standard equipment for workers at the Swiss Nuclear Center at Würenlingen (47).

In the nuclear safeguards device shown in Fig. 10 the same principles are used to record the neutron energy spectrum and flux at nuclear reactors as a function of time for future readout (48). Once the flux level and neutron energy spectrum are known, the uses to which a reactor is being put can be recognized. In the sealed unit sketched in Fig. 10 fission plates containing  $^{235}\text{U}$ ,  $^{237}\text{Np}$ , and  $^{232}\text{Th}$  give fission fragments from neutrons of different energy, which are recorded on

a moving detector tape to provide time resolution. A  $^{252}\text{Cf}$  fission source records the tape speed and its gradual decay in intensity provides an absolute time calibration.

Heavy particle dosimetry has provided what is probably the most esoteric dosimetry application to date: the measurement of the heavy ion cosmic-ray dose received by the Apollo 8 and Apollo 12 astronauts. This was done by simply etching the astronaut helmets, which are made of Lexan plastic, to reveal particle tracks (49, 50) such as those shown in Fig. 11. This measurement is particularly relevant since the ionization rate necessary to produce a track in Lexan is close to that for which a nuclear particle crossing certain

cells will kill the cell with near unit probability. Thus the track measurements can be related directly to total cell damage. These measurements indicate that a substantial number of brain cells ( $\sim 0.1$  percent) might be destroyed on an extended space mission (50). More routinely, Apollo astronauts wear film packets that include plastic dosimeters (51).

In addition to measuring cosmic rays and thermal neutrons, track detectors can be used to monitor doses of energetic fundamental particles. The technique used here is to observe interaction secondaries produced by the high-energy particles, most frequently induced fission events. This dosimetry can be useful in such diverse uses as

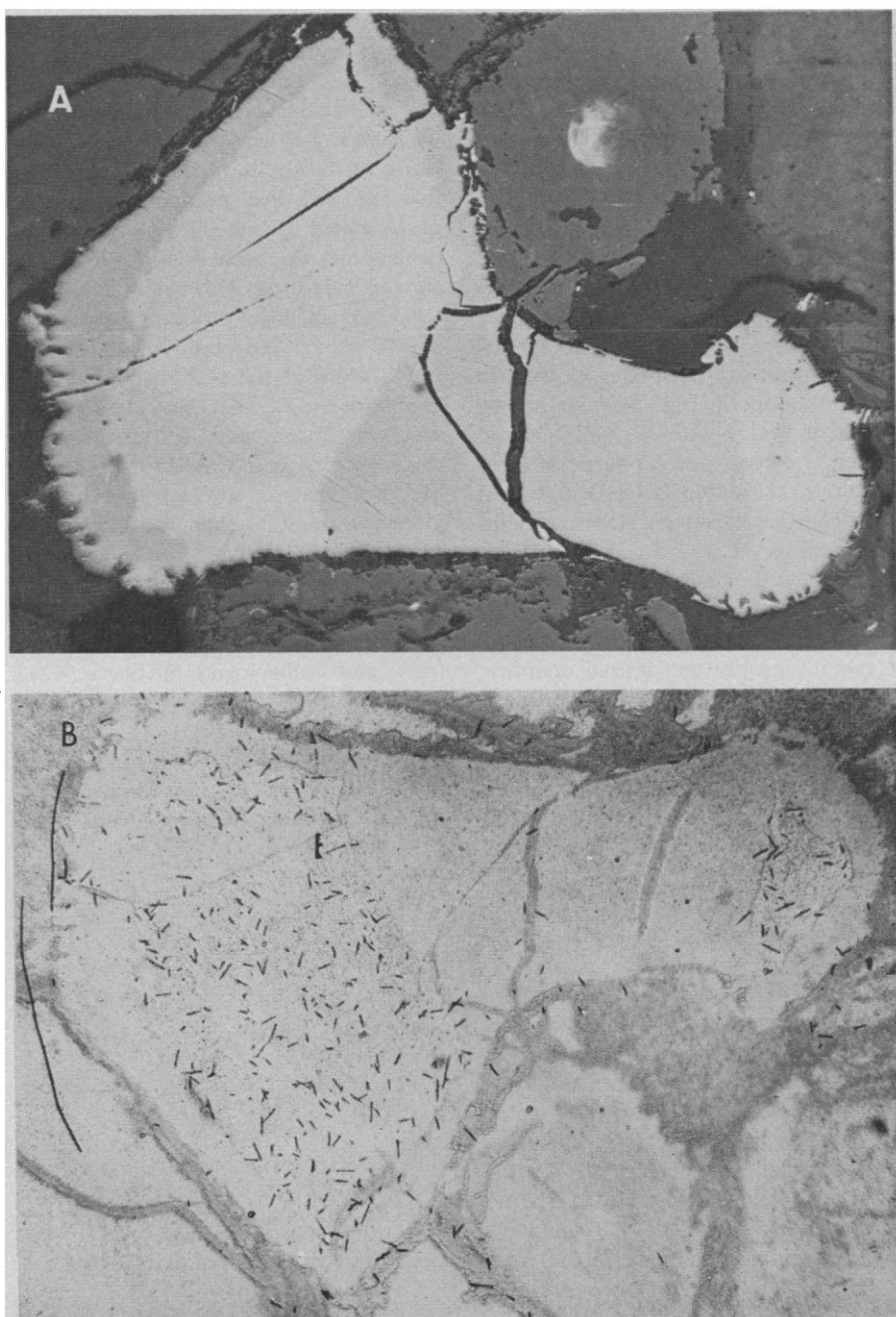


Fig. 13. A plastic sheet was neutron-irradiated while held against a polished rock section (A) and then etched. The Lexan "print" (B) allows ready differentiation of minerals and their uranium contents. Alpha and fission tracks provide distinct textures in different minerals. The track distribution shows that the light phase, rutile, contains 1.1 parts per million of uranium (64). The maximum fission track length is approximately 18 micrometers. [Courtesy of *Atomic Energy in Australia*, Coogee, Australia]

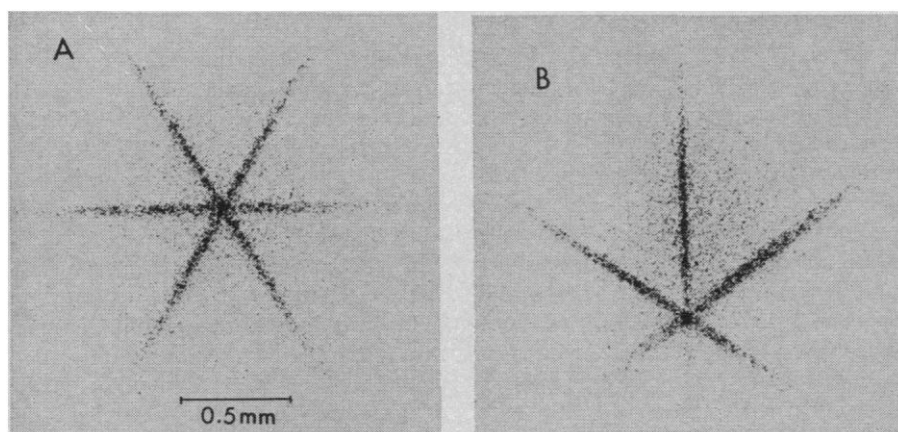


Fig. 14. Channeling through small molybdenum crystals serves to display their orientations;  $^{241}\text{Am}$  alpha particles penetrate the crystals preferentially along low index planes and directions and display the crystal symmetries in a cellulose nitrate detector: (A) a  $\langle 111 \rangle$  orientation; (B) a  $\langle 210 \rangle$  orientation. Based on the technique outlined in (66). [Courtesy of J. Mory]

monitoring beams at high-energy accelerators (39) or in detecting the bombardment of objects on the moon by high-energy cosmic-ray nuclei (52).

The same sensitivity to alpha particles that allows  $^{222}\text{Rn}$  from uranium ore to be measured is useful in monitoring the exposure of mine workers to alpha particles (53). The primary source of damaging alpha radiation is thought to be the decay of daughter products of  $^{222}\text{Rn}$  trapped in aerosol particles that are inhaled and trapped in the lungs. In Fig. 12A is shown an active device with a small fan and a filter to extract aerosol particles from the air in a uranium mine and record the alpha decays on an adjacent cellulose detector (54). In Fig. 12B is shown a still smaller, passive device to record  $^{222}\text{Rn}$  alpha decays (55). The former device has the advantage of recording only the desired quantity; the latter has the merits of simplicity, ruggedness, and light weight.

A more extensive bibliography of dosimetry applications is available from the authors (23). A review containing 200 references is given in (55a).

#### Miscellaneous Track Applications

Further diverse applications involve chemical analysis and metallurgical techniques. Chemists and geochemists make use of track detectors not only to map chemical distributions, as noted earlier, but also to perform quantitative chemical analysis for fissionable nuclei or nuclei susceptible to  $(n, \alpha)$  reactions (11, 30, 56–65). Because the track densities obtained depend lin-

early on the neutron dose used to induce nuclear reactions, the use of high fluxes allows very tiny quantities of fissionable nuclei to be detected and measured, the limitations on sensitivity being set by the purity of the detectors that are used. For uranium, an element of immense geochemical importance, concentrations below 1 part per billion can be measured with reasonable ease (30, 56) and local groupings of as few as  $10^7$  atoms are readily recognizable (11). Work at the National Bureau of Standards (57, 58) has shown that uranium, boron, and nitrogen can be measured by track detectors with an accuracy that is as high as that of more conventional chemical techniques. This technique has been used to measure the uranium concentrations of water in its natural environment (59), biological material (60), fossil bones (61), and archeological artifacts (62).

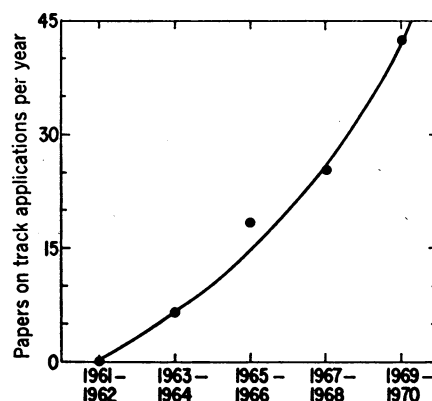


Fig. 15. Rate of publication of applied track etching work as a function of time. Data for this graph were current as of June 1971. Data points are 2-year averages. The curve was drawn by hand.

One delightfully simple and useful geochemical technique involves the use of Lexan "prints," which are etched plastic detectors that were pressed closely against a rock section during neutron irradiation. As displayed in Fig. 13, different mineral grains are readily distinguished on the basis of the different textures of the adjacent Lexan after etching (63, 64). Here both  $(n, \alpha)$  and  $(n, \text{fission})$  effects appear, with most of the contrast supplied by the differing boron, oxygen, and lithium concentrations of the various minerals.

Metallurgy is another area in which tracks are useful. Tracks can serve as above as an analytical tool in the measurement of diffusion constants at trace concentrations (66) or in crystallographic applications such as orienting crystals by means of channeled particles (67) and detecting atomic and structural imperfections (68). Figure 14 shows an example of a channelograph—the etched tracks of  $^{241}\text{Am}$  alpha particles channeled preferentially along certain crystal planes and directions through crystals of tungsten 10 micrometers thick. The crystal symmetries that are displayed by etched tracks in a sheet of cellulose nitrate make it possible to infer crystal orientations readily.

A bibliography on chemical applications of particle track etching is available from the authors (23).

#### Concluding Remarks

The work we have described here began at a time when three of the authors (R.L.F., P.B.P., and R.M.W.) were at the General Electric Research Laboratory and the other two (H.W.A. and S.C.F.) were at the General Electric Vallecitos Nuclear Center. Although the use of particle track etching has been worldwide for some years now (69), we believe that the initial rapid discovery and utilization of the technological applications of track etching came about because of the conscious, deliberate juxtaposition within one organization of workers who wanted to do scientific work and those who wished to use scientific results. Most of the initial scientific applications of track etching came from the Research Laboratory and most of the initial technological uses were conceived and explored at Vallecitos Laboratory, making use of the new opportunities revealed by the new science.

Where do particle track applications go from here? Figure 15 gives us a hint. Papers on the applications of track etching have been published at a rate that has doubled every 2 to 2½ years over the last few years. It seems likely that many new applications that have just begun to be explored will develop into new areas of usefulness.

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