Track-recording solids

Simple dielectric solids, which reveal through chemical etching the tracks left by fast-moving nuclei, are being used as particle detectors in a growing number of scientific and technological areas.

Steven P. Ahlen, P. Buford Price and Gregory Tarlé

Since the beginning of the solar system, natural particle detectors have been recording the passage of charged particles from the sun and cosmic rays. Now, in addition to developing the latent images of these fossil trails of damage in solids and learning about the nature of ancient radiation, we are creating new and more sensitive detectors of a similar kind. These detectors, which are finding a wide variety of applications, take advantage of the fact that a highly charged particle penetrating any nonconducting solid leaves a submicroscopic trail that can be chemically amplified. The increased chemical reactivity of the trails of radiation-damaged material is the basis for the so-called etched-track process,[1-5] by which we make the particle tracks large enough to measure in an optical microscope. As we will see, there is sufficient information in the tracks to allow us to determine a particle's charge and velocity.

We begin this article by describing the mechanisms by which particles produce trails in dielectric solids and the techniques that we use to measure the tracks and identify the responsible particles. Then we discuss the application of these solid-state particle detectors in space physics, nuclear physics, fission track dating, nuclear imaging and various technologies. Finally, we will cover some recent advances in high-resolution particle identification. We hope that some readers who have not previously heard of nuclear tracks in solids will conceive of entirely new applications to problems that interest them.

Steven P. Ahlen and Gregory Tarlé are assistant research physicists in the Department of Physics and in the Space Sciences Laboratory at the University of California, Berkeley. P. Buford Price is professor of physics and director of the Space Sciences Laboratory.

Track production and detection

As we have said, ionizing radiation passing through a dielectric solid leaves a trail of damage that is chemically reactive. It is possible to view this latent image with an electron microscope without developing it chemically. From such direct observations and other indirect observations we know that the latent image consists of atomic or molecular (not electronic) defects that are concentrated within a few nanometers of the particle's trajectory.[1] Among the consequences of the small lateral extent of this chemically reactive region are

- great uniformity in the rate of radiation damage by each particle of interest, a reproducibility that contributes to our ability to identify nuclear particles
- insensitivity of the chemical reactivity of one track to nearby tracks and to overall radiation damage
- ability of the solid to record highly ionizing particles without recording lightly ionizing particles
- ability to record more than 10^{11} tracks/cm^2 that can be resolved in an electron microscope
- technological applications such as microfiltration and ion microscopy.

Irradiation experiments with ions of various charges (Z) and velocities ($\beta \equiv v/c$) and with electrons and gamma rays indicate that tracks are formed differently in inorganic solids and in organic polymers. The "ion explosion spike" model applies to minerals and glasses. Here an energetic particle forms a latent track by producing a high enough concentration of positive ions that the resulting electrostatic pressure exceeds the mechanical strength of the solid. After an explosive ejection of ions from the trajectory of the particle, the stressed region, about a nanometer in diameter, relaxes elastically, leaving a long-range strain field that can be seen by electron microscopy (see figure 1) and chemically etched. The model explains why tracks do not form in conductors; why tracks form most easily in solids with low mechanical strength, low dielectric constant and close interatomic spacing; and why chemical reactivity depends on the primary ionization rate rather than on the total energy-loss rate.

The validity of the ion-explosion-spike model receives additional support from recent experiments in which atoms are "sputtered" or ejected from

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Track-recording solid (left) shows macroscopic etched tracks of 100-GeV Fe^{56} nuclei brought to rest in a stack of five slabs of allyl diglycol polycarbonate doped with 1% of dioctyl phthalate. Careful examination of the plastic slabs shows numerous examples of nuclear fragmentation. Etching for one month in a warm sodium hydroxide solution removed about 0.75 mm of material from the surfaces and caused the cone-shaped track etch pits to develop. Transmission electron micrograph (above) shows elastic strain fields around tracks left by fission fragments penetrating mica.[1] Hydrofluoric acid or hot sodium hydroxide etches the damaged regions preferentially, enlarging the holes. The longest tracks visible in the photograph are about 0.2 micron in length. The area of the micrograph is about the size of a single silver halide grain in nuclear emulsion. Figure 1

surfaces of dielectric solids when they are bombarded with ionizing particles. Previously, sputtering was thought to involve a cascade of atomic collisions, with the maximum sputtering yield (the average number of atoms ejected per incident ion) occurring at an ion energy of only a few keV per atomic mass unit, for which the energy loss rate by *atomic* collisions is a maximum. New experiments[6] with beams of fluorine ions of various energies show that the sputtering yield depends on the fluorine ion energy in the same way as does the primary ionization rate, and reaches a maximum at a fluorine energy of about 300 keV/amu. This is expected if the ion explosion spike mechanism is responsible for ejecting atoms by Coulomb repulsion and for exceeding the sputtering yield expected from an atomic collision model by a factor of about 10^3.

Polymers are much more sensitive to ionizing radiation than are most inorganic solids. The ion explosion spike is quite unimportant compared with the scission of long-chain molecules, which occurs as a result of the decay of excited electrons and typically requires only 1 or 2 electron volts. Chain scission results in molecules that are smaller and of increased chemical reactivity. Radiation also causes crosslinking—the generation of primary bonds between polymer chains—which locally decreases the chemical reactivity. In some polymers irradiation principally causes crosslinking; in others it principally causes chain scission. Radiation chemists use the quantities G(scission) and G(crosslink), defined respectively as the number of chain scissions per 100 eV of deposited energy and the number of crosslinks per 100 eV, to measure sensitivity to gamma rays or high-energy electrons.

We qualitatively define the etched-track sensitivity of a solid as the minimum value of Z/β for a nuclear particle to produce an etchable track in that solid, $(Z/\beta)_{min}$. Solids with the smallest $(Z/\beta)_{min}$ are the most sensitive. This simple ratio is useful because the radiation damage rate along the trajectory of a particle is related to its primary ionization rate, which is proportional to $(Z/\beta)^2$ times a factor only weakly dependent on β. For the polymers in which both tracks and overall radiation damage due to gamma rays have been studied,[7] $(Z/\beta)_{min}{}^2$ is proportional to $[G(\text{scission})]^{-1}$, which means that the minimum value of the primary ionization rate for track production is proportional to the energy per scission in gamma irradiation. $(Z/\beta)_{min}{}^2$ does not correlate at all with G(crosslink). Thus, our searches for new applications and for polymers that are even more track-sensitive ally us with chemists who synthesize polymer "resists" with high G(scission) from which masks for very large integrated circuits are made by electron beam irradiation. Most of the known track-sensitive polymers have sensitivities, $(Z/\beta)_{min}$, between about 5 and 100; these are intermediate between nuclear emulsion, with $(Z/\beta)_{min} = 1$, and minerals and glasses, with $(Z/\beta)_{min}$ ranging from about 150 to 450.

A polymer that is specifically tailored for the imaging, automated locating, or accurate measuring of nuclear particle tracks must have several properties:

- Sensitivity to particles of interest and insensitivity to lighter particles and photons. Ideally one would like to be able to tune the sensitivity of a specific material with a trace additive or a minor adjustment in chemical etchant.
- Optical clarity. Precise measurements of the tracks, automated scanning, and imaging require that the surfaces be highly transparent after etching.
- Uniform composition and morphology. Regions of differing composition or polymer architecture lead to fluctuations in the rate of chemical etching, causing rough surfaces that impair resolution.
- Dominance of scission or chain-breaking over crosslinking upon irradiation. To produce etched tracks, the etch rate along the particle's trajectory must increase, not decrease.
- Time-dependent response to a charged particle and latent-image stability at ambient temperature.

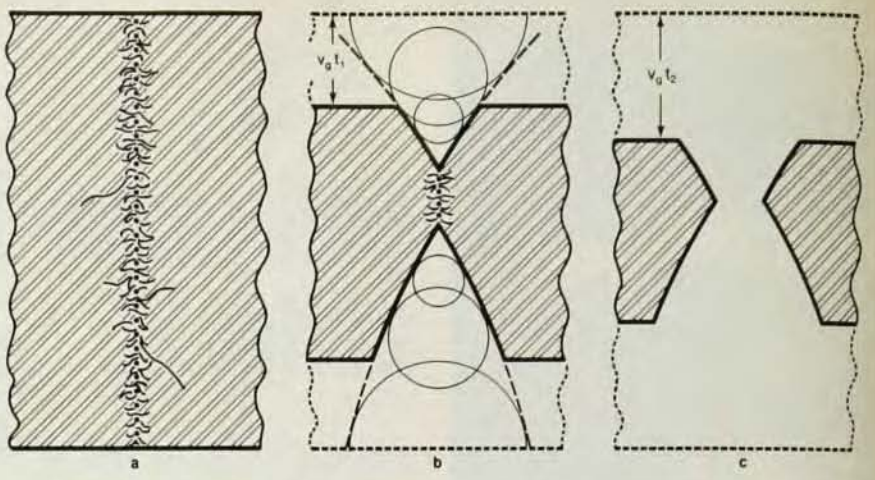
In concluding this brief discussion of tracks in polymers we should note that heavy ions from the LBL Bevatron appear to produce tracks in rat corneal tissue,[8] a sobering fact that should be considered when subjecting humans to medical treatment by heavy ions or

exposing humans to cosmic rays in space.

Figure 2 illustrates the quantitative basis of the etched-track technique. In (a) an energetic nuclear particle has penetrated a track-recording solid, leaving a trail of molecular or lattice defects whose density falls off radially by several orders of magnitude within about 4 nm of the trajectory.[1] Although it is possible to observe the latent track by transmission electron microscopy, virtually all scientific and technological applications depend upon the ability to find an appropriate chemical etchant—a reagent that breaks bonds solely at the liquid–solid interface and enlarges the track in a controllable way.

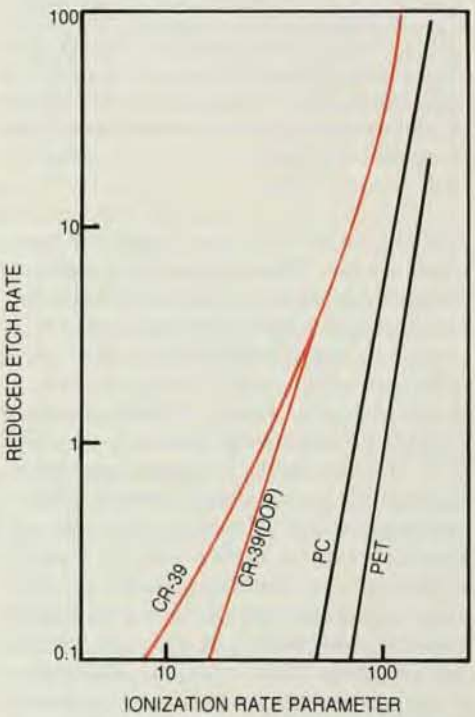
It is very fortunate that the track-etching process can be quantitatively specified by only two parameters: the general etch rate, v_g, which ideally is constant at all free surfaces for a given etchant and etch temperature; and the track etch rate, v_t, which gives the rate of etching at the point where the latent track reaches the surface and is an increasing function of Z/β. Figures 2(b) and 2(c) indicate how the etched-track process works in an isotropic solid. A particle enters the top of the solid with initial velocity β_i and ionization rate parameter Z/β_i, slows down as it produces a latent track, and emerges from the bottom with a lower final velocity β_f and, therefore, a higher ionization rate parameter Z/β_f. The slight curvature of the conical etch pits in figure 2(b) quantitatively reflects the changes in Z/β as the particle slows down, in the same way as the bow wave of an accelerating boat displays the history of the boat's speed. The etch rate v_g plays the role of the water wave speed, and v_t plays the role of the boat's speed.

To determine the charge and speed of an unknown particle we first establish, by calibration with known particles at a heavy-ion accelerator, the dependence of Z/β on the etching-rate ratio, s (v_t/v_g), and the decrease in β with depth of penetration. Examples of the former are given in figure 3. Then we measure the dimensions of the etch pits at two or more locations such as the top and bottom of the solid in figure 2(b). Often it is sufficient to measure a small number of parameters such as the minor axis of the elliptical mouth, the depth of each etch pit and the angle of inclination to the vertical. For a conical etch pit with half-cone-angle θ and walls with nearly zero curvature (implying an approximately constant Z/β), the etch-rate ratio is given by the simple relation $s = \operatorname{cosec}\theta$. Thus measurement of s at two points on the trajectory of a single particle gives us two values of Z/β. Our knowledge of the decrease in β between the two points leaves us with two equations, which we solve for Z and β.



Sketch of a nuclear track in a dielectric solid before and after etching. Particle entered from above. By measuring the etch pit profiles, which reflect the etch rate along the track and the general etch rate that acts normal to every exposed surface, one can determine the charge and velocity of the nuclear particle. Figure 2

Extraterrestrial recordings. The lunar surface, meteorites, and other objects in space have been irradiated by charged particles from a variety of sources in the Sun and the Galaxy.[5] The essential permanence of latent tracks in most natural solids at ordinary temperatures opens up two complementary lines of study of the past: If the history of an extraterrestrial solid is known and simple, then one can use the fossil track record to study highly ionizing particles emitted by the ancient Sun, to study cosmic rays at various times in the past, and even to study the short-lived transuranic elements that decayed by spontaneous fission soon after the solar system formed. Or, if the sources of tracks are well understood, then the study of the distribution and intensity of fossil tracks leads to knowledge of the age, dynamics, and thermal history of the meteorites, of the lunar crust, and of the Earth's crust and sea-floors.



Calibration curves showing the reduced etch rate ratio $(v_t/v_g - 1)$ as a function of the ionization rate parameter (Z/β) for polyethylene terephthalate (PET), Lexan polycarbonate (PC), high-quality allyl diglycol polycarbonate (CR-39) and CR-39 doped with dioctyl phthalate (CR-39(DOP)). Figure 3

We mention only one of the numerous applications of fossil tracks to solar system history. Figure 4 shows etched tracks of energetic iron nuclei from solar flares in a 0.15 mm-diameter olivine crystal from a carbonaceous chondrite—a class of meteorites whose members are much studied because of their primitive, unmetamorphosed structure. Detailed analysis of this and other crystals shows that the irradiation occurred about four billion years ago, before the individual grains were compacted into a meteorite, and that the energy spectrum of iron nuclei emitted in flares by the ancient Sun is similar to the average spectrum in recent flares. From statistical analyses of densities, gradients and anisotropies of solar flare tracks in olivine grains and from auxiliary rare-gas evidence, it has been possible to construct a plausible model for the rate of accretion of matter during the formation of the solar system: coalescence into swarms of centimeter- to meter-sized bodies, followed within about 10^5 years by coalescence into kilometer-sized and larger objects.[9]

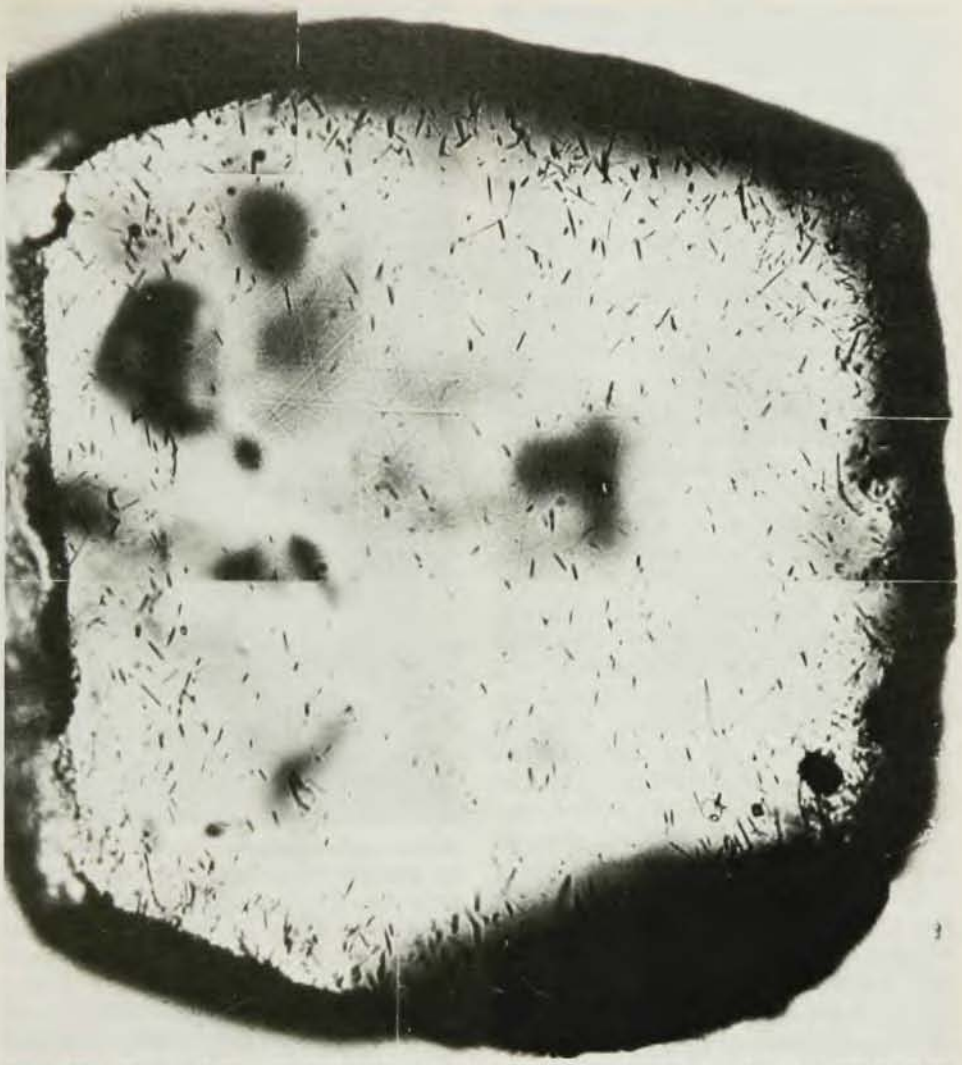
Fission track dating. In terrestrial solids, which are well shielded from external radiation, the dominant source of tracks is the spontaneous fission of uranium-238, present as a trace-element impurity. Although the fission

rate may seem incredibly small—the decay rate for spontaneous fission is about 10^{-16} per year per atom—the concentration of uranium-238 is rarely lower than about 10^{15} atoms per cubic centimeter, so that tracks accumulate over geologic time at a readily measurable rate, which has led to the extraordinarily productive fission track dating method.[1] The density of tracks depends on the product of the uranium concentration in the sample and the time since it began to record the fission events. In practice we determine the uranium concentration by irradiating the sample with thermal neutrons, which induce fission in uranium atoms. The number of new tracks produced by the fission products is of known proportion to the number of uranium atoms present. (The existence of natural fission tracks is a case where a geochronologist's signal is a physicist's noise, for if spontaneous fission did not occur, particle physicists might be able to look for such phenomena as spontaneous proton decay and coherent neutrino scattering by examining tracks of recoiling nuclei in minerals deep underground.)

Because fission tracks are erased at a well-defined temperature for each particular mineral (for example, 100 °C in apatite) one can use the apparent fission-track ages as a function of distance from a heat source to measure the thermal history of regions. Examples are the rate of sea-floor spreading, about 1 centimeter per year, and the rate of uplift of mountains, which ranges from about 10^{-2} centimeter per year for the Wasatch range in Utah to the astonishingly high current value of about 1 centimeter per year for the Himalayas, according to Charles W. Naeser.

Space physics. Although the existence of cosmic rays heavier than iron was first established by analyses of fossil tracks in meteorite minerals,[1] a reliable charge identification system for ancient tracks of very heavy nuclei in minerals has proven elusive—a pity in view of the enormous collecting power for hypothetical particles of high ionization rate. Glasses and plastic sheets, being amorphous and isotropic, fare much better. It was the quantitative study of tracks recorded during the exposure of glass and plastic detectors carried on spacecraft and in balloons that led to the discoveries of transuranic cosmic rays,[1] of energetic heavy nuclei in the Earth's radiation belt,[10] and of the preferential emission of heavy rather than light nuclei in solar flares.[1]

Nuclear physics. Experiments with solid track detectors have greatly enriched our understanding of the fission process as well as of relativistic heavy ion reactions. Track-recording solids have led to discoveries of a variety of phenomena including fission into three fragments,[1] numerous far-transuranic nuclides,[1,11] very short-lived isomers that spontaneously fission from the outer well of a double-humped fission barrier,[1] and the most neutron-rich nuclides ever produced—C^{20} and F^{27} (reference 12).



Four-billion-year-old fossil tracks of energetic iron nuclei in a 0.15 mm olivine crystal from the deep interior of a carbonaceous chondrite meteorite. The source of the irradiation was solar flares that occurred before the crystal was buried inside the meteorite. Studies of such tracks give the energy spectra of ancient flares and indicate a scenario for the accretion of matter at the beginning of the solar system. (From reference 24.) Figure 4

Nuclear imaging. Everyone is familiar with the versatility of visible-light and x-ray photography. But nuclear photography, using track-recording solids, also has an extraordinary number of applications. Each picture element, or pixel, is a single track that can be amplified by etching to any desired size from about 10 nm on up. Depending on the concentration of pixels and the extent of etching, one can produce an image that is visible to the naked eye, that can be printed through as though it were a photographic negative, that can be examined microscopically with very high spatial resolution, or that can be processed by computer techniques. One of the most successful and beneficial applications is heavy-ion radiography,[13] which uses a heavy-ion beam of a few hundred MeV/amu for imaging minuscule variations in tissue density to locate tumors that would be invisible to x rays. The superior contrast in such images is a result of the extremely well-defined range of monoenergetic heavy ions.

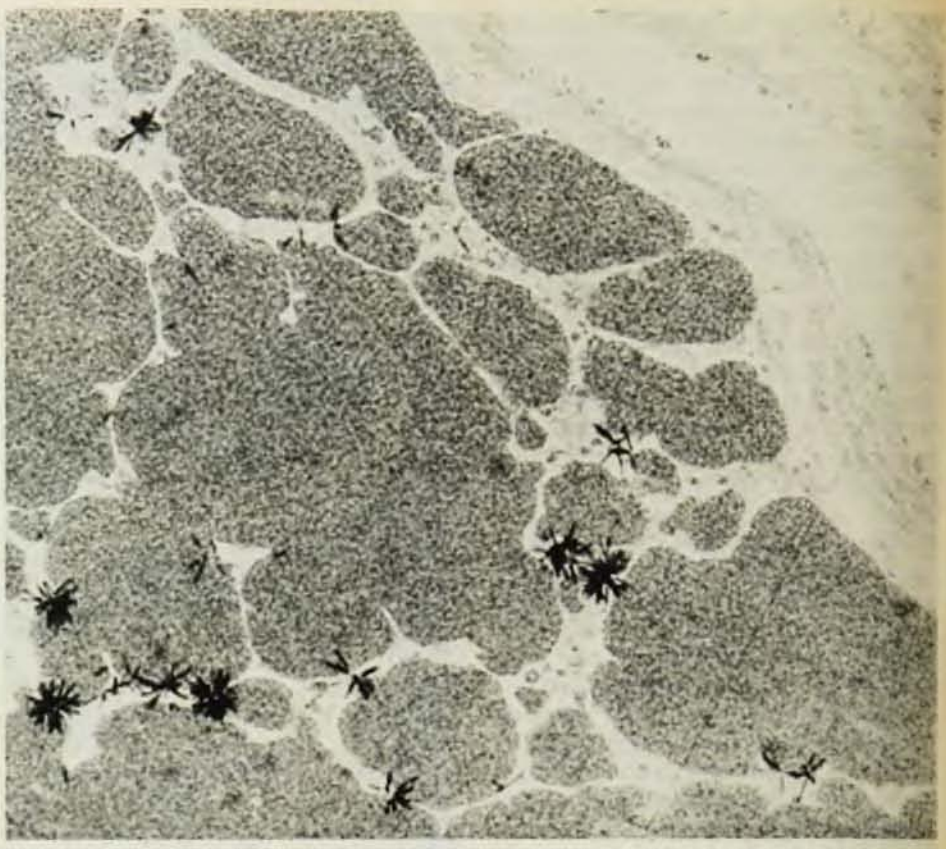
Microscopic mapping of trace element distributions, using mica or polycarbonate films, has been a boon to geochemists, radiobiologists, and workers in many other fields. To produce an image one initiates a nuclear reaction such as $n + U^{235} \rightarrow$ fission products, or $n + B^{10} \rightarrow He^4 + Li^7$, which produces in an adjacent film tracks whose locations map out the U^{235} or B^{10} distribution in the sample. Alternatively one may use a reaction such as $He^3 + Pb^{206} \rightarrow 3n + Po^{206}$, which leads to a radioactive product that decays by emitting a heavily ionizing particle. In the latter example Pb can be micromapped without interference from other elements by placing an alpha-sensitive solid against the sample a couple of days after the irradiation and leaving it

until all of the Po^{206} (8.8-day half-life) has decayed by alpha emission.

Heavy-ion microscopy using plastic as the recording medium has enormous potential. As we will see, this lensless microscopy, due to Cornelius A. Tobias,[14] exploits the extremely small variation of range of a monoenergetic beam of heavy ions in matter. Nevertheless, the small amount of range-straggling does limit the resolution to a few hundred angstroms. Although electron microscopy has better resolution, heavy-ion microscopy allows one to study thick, living cells and other hydrated objects directly, providing information on small variations of density within the cell and permitting the direct examination of aberrations in chromosomes. The object to be imaged is placed on a plastic sheet and irradiated with a beam of low-energy heavy ions that barely penetrate the object and stop at different depths in the underlying plastic depending on the density of matter traversed. Etching the plastic produces an image of the stopping points of the beam. This image is a density replica of the object and can be examined in a scanning electron microscope.

An example is a recent study of the incorporation of radioactive material in tissue.[15] Rats inhaled an aerosol of UO_2 enriched in U^{234} and a week later 5-micron-thick tissue sections were prepared. To study the distribution of uranium-234, which has a half-life of 2.4×10^5 years, researchers kept the sections in contact with allyl diglycol polycarbonate plates for a month, during which time some of the U^{234} produced alpha tracks in the plates. Then they irradiated the tissue sections, still in contact with the plastic, with a parallel beam of 1 MeV alpha particles ($10^9/cm^2$) to produce an image of the density variations in the tissue. After removing the tissue and etching the plates, the exact locations of the radioactive UO_2 particles could be seen, superimposed on the tissue image. The photograph in figure 5 is the result of applying the technique in conjunction with alpha-particle autoradiography to determine the microdistribution of alpha-emitting uranium-234 in a histological section of rat lung tissue. Researchers at Bristol and other laboratories are studying the tracks of alpha-particles from natural polonium-210 (from tobacco) in lung tissue of cigarette smokers. By using a polymer that can be pre-etched to remove background tracks without roughening the surface, it should be possible to detect activities as low as 1 track/cm^2.

Several laboratories are using plastic detectors to study charged particles emitted in fusion reactions. Particularly beautiful is the zone-plate coded-imaging technique for microscopic diagnosis of laser fusion targets.[16] A coded image of the fusion region is obtained by using a Fresnel zone-plate aperture to cast a shadow onto a sensitive track-recording solid. After etching, the transparent detector will focus incident laser light into multiple, well-separated, reconstructed images, as shown in figure 6. The large collecting power of the zone-plate camera compared with a pinhole camera allows one to reconstruct an image of the burning thermonuclear fuel with a resolution approaching 1 micron, even when only about 10^8 particles are emitted.



Alpha autoradiograph showing numerous tiny, black, starlike clusters of alpha tracks emanating from particles of uranium dioxide rich in U^{234}, superimposed on an ion image of rat lung tissue.[15] The sample in the picture is 0.6 mm wide. (Courtesy of Donna J. Gore and Terrence J. Jenner.) Figure 5

Technological uses. The idea of a molecular sieve occurs to almost everyone who sees figure 2(c). The Nuclepore Filter, with a multitude of uses, was an almost immediate spinoff of the discovery of track etching.[1] This filter, made by irradiating a polycarbonate film with fission fragments and etching it, can have holes of any desired concentration and diameter from about 30 nm to 10 microns, depending on irradiation time and etch time. Aside from obvious applications such as in biomedicine and draft beer production, a product with straight holes having a length-to-diameter ratio comparable to that of deep oil wells is bound to have more

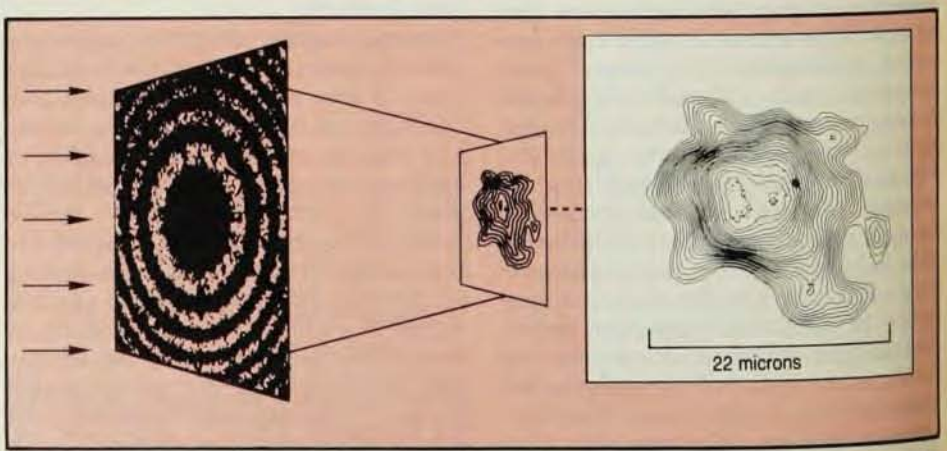


Diagram of zone-plate coded-imaging recording technique. After irradiation through a zone-plate aperture, the plastic detector is etched and used to bring laser light to a focus in various planes. The actual reconstructed third-order alpha-particle image shown here has a resolution of about 3 microns. (Courtesy of Natale M. Ceglio) Figure 6

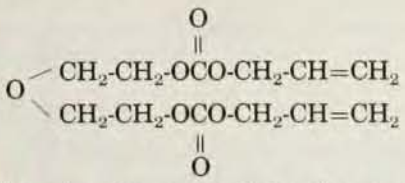
exotic applications. For example, by using a sheet with a single, submicron hole as a partition between two conducting fluids, one can measure the size, shape, and mobility of viruses, sea-urchin sperm, and other submicroscopic organisms.[17]

The storage density in a nonconducting magneto-optic film can be greatly increased by irradiating the film with heavy ions and etching the tracks. Magnetic domain boundaries will then go preferentially though the etched tracks, thereby lowering the total magnetic field energy and permitting a much higher density of domains and of stored information than in a nonirradiated film.[18]

Sensitive plastic detectors are being used to measure low levels of fast neutrons by recording recoil proton tracks, and to detect very low concentrations of radon gas in soil and air by recording alpha-decay tracks. Careful measurements of the spatial and temporal distribution of Rn^{222}, a 3.8-day daughter in the U^{238} decay chain, indicate the locations of uranium ore deposits and may assist in earthquake prediction. Perhaps even more important is the use of plastic detectors[19] to measure the very low levels of radon, typically about 1 picocurie/liter, in homes. The simple act of conserving home fuel by reducing ventilation has been shown to increase the radon concentration. The U.S. Environmental Protection Agency estimates that reduction of ventilation in the average American home by a factor of two would increase the incidence of lung cancer by 10 000 to 20 000 cases per year as a result of increased exposure to radon and its decay products. Fortunately, ways of increasing energy efficiency of homes without increasing radon concentration are being developed, and plastic track detectors will continue to play a key role in this kind of research.

A remarkable plastic

The many scientific and technological applications of track recorders has stimulated efforts toward finding better recording solids. The discovery in our laboratory[20] of the remarkable track-recording properties of allyl diglycol polycarbonate, a polymer with the trade name CR-39, was the result of a rational search for a plastic sufficiently uniform to permit the resolution of adjacent elements and perhaps even adjacent isotopes of relativistic heavy nuclei. Unlike Lexan polycarbonate and other thermoplastics extruded at high temperature, CR-39 is made by adding about 3% of a heat-sensitive initiator to the monomer, which is then cast and heated. The heating initiates polymerization and crosslinking of nearby molecules of the allyl diglycol carbonate monomer shown below:

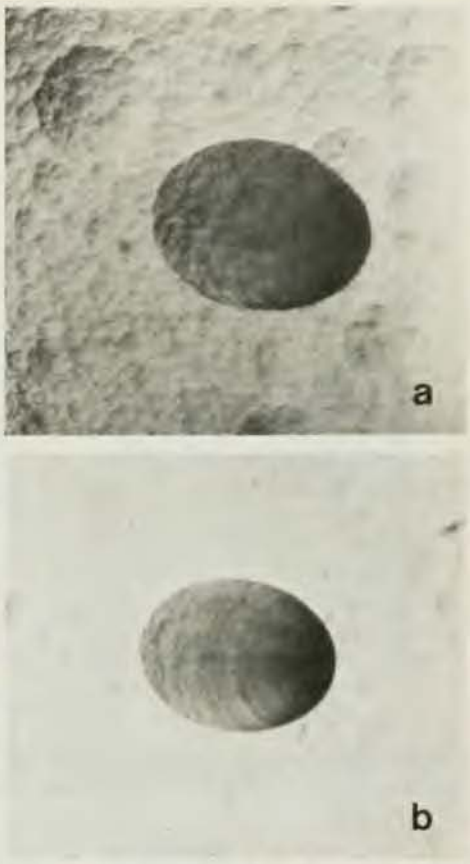


Polymerization proceeds by the opening of C=C double bonds and the connecting of all the monomers in a gigantic, crosslinked network.

The resulting product is the most sensitive track-recording solid known. It is capable of extremely high charge resolution, and etches far more uniformly than do other polymers, making possible new scientific applications that will be discussed in the remainder of this article.

By adding to the monomer a small quantity of a class of organic compounds typified by dioctyl phthalate (DOP), we have been able to produce polymer detectors with greatly improved post-etch optical properties.[21] By analogy with the nomenclature for Li-doped semiconductor detectors, the doped CR-39 detectors are designated CR-39(DOP).

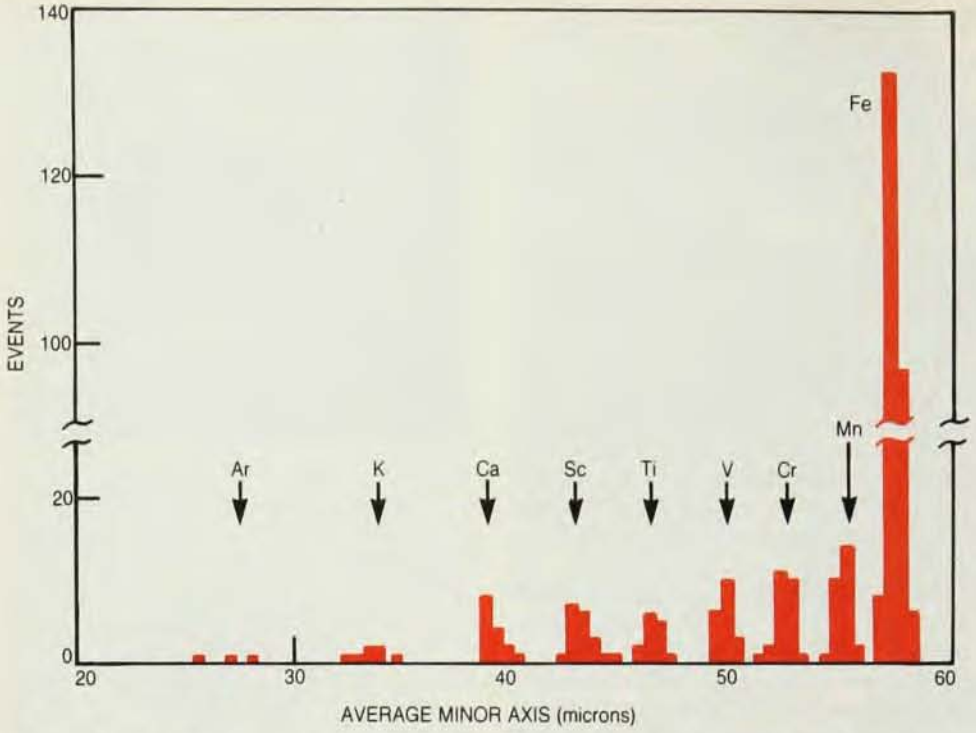
Lexan polycarbonate, one of the most widely used track-recording solids, is too sensitive for some applications and develops a rough surface and irregular tracks during etching. Cellulose nitrate, though much more sensitive than Lexan, develops extremely rough surfaces and etch-pits and suffers from the fading of latent-images. Carefully made CR-39, though acceptable for many applications, still has one drawback. If etching is continued until the track diameters reach several tens of microns, the surfaces become rough, suggesting that the polymer contains nonrandom structural inhomogeneities on roughly the scale of the wavelength of light. We found[21] that by adding about 1% of a plasticizer such as dioctyl phthalate to CR-39 monomer, the resulting sheet remained beautifully smooth and the etch-pits had superb optical quality even after prolonged etching (see figure 7). To account for the striking effect of such a small concentration of plasticizer, we note that CR-39, a highly crosslinked thermosetting plastic, suffers from inhomogeneities known to the polymer chemist as crosslinked clusters. Regions of high crosslink density, which are more resistant to chemical attack, are attached to each other by regions of lower crosslink density. As chemical etching proceeds, these inhomogeneities manifest themselves as a general surface roughness. It appears that molecules of dioctyl phthalate, or other large phthalic acid esters, interpose themselves between the polymer chains and inhibit the formation of crosslinked clusters, with the consequence that the general etching rate, v_g, is highly uniform.



Interference micrographs of elliptical etched track mouths at the top surface of (a) undoped CR-39 and (b) CR-39(DOP). The advantage of the image in (b) for automated measurements is obvious. The greater transparency of etched CR-39(DOP) enables successive measurements to be made on a superimposed stack.[21] Figure 7

High charge resolution

A charged particle slows by making numerous energy transfers to electrons in the medium it traverses. The infrequent "close" collisions lead to high-energy electrons called delta rays; the frequent "distant" collisions lead to excitations and ionizations in which electrons are left with very little kinetic energy. The most fundamental limitation to the charge resolution of a detector that measures the energy loss of penetrating charged particles is the fluctuation of the amount of energy that produces the signal in the detector. Because the variance of the signal is proportional to the maximum energy transfer to which the detector is sensitive, ϵ_m, a detector with a low ϵ_m suffers less from energy loss fluctuations than does a detector with a high ϵ_m. Plastic scintillators suffer worst because they respond preferentially to the infrequent high-energy delta-rays, which show the largest fluctuations; semiconductor detectors are intermediate, responding to total energy loss; and dielectric track detectors, because their chemical reactivity is increased only in a region (about 4 nm across) associated mainly with excitations and delta-rays



Data showing charge resolution of CR-39(DOP) for relativistic heavy ions.[21] For a given thickness in g/cm^2, the resolution of this extraordinary plastic exceeds that of any other energy-loss detector. Figure 8

with ϵ_m about 300 eV, are nearly immune to energy-loss fluctuations. This leads to the prediction[20] that dielectric track detectors should be capable of higher charge-resolution than any other type of detector, including semiconductor diode devices, the current favorite whenever small size and extreme expense can be tolerated.

Figure 8 shows some of the results of a recent experiment with a 1.86 GeV/amu beam of iron-56 nuclei at the Lawrence Berkeley Laboratory Bevalac in which we demonstrated that CR-39(DOP) has a charge resolution superior to that of a silicon detector.[21] We placed ten sheets of CR-39 behind a polyethylene slab that caused about 60% of the iron nuclei to fragment as they passed through. The velocity loss was sufficiently small that we could neglect velocity dispersion effects. The odd-numbered sheets (number 1 being closest to the polyethylene) had 1% dioctyl phthalate added and the even-numbered sheets did not. Following exposure, we etched all ten sheets in a sodium hydroxide solution until about 70 microns of material was removed from each side of each sheet by general etching.

We studied the charge resolution of CR-39(DOP) by measuring the minor axes of the elliptical etch-pit mouths in the top and bottom of some odd-numbered sheets. Because of the uniform transparency of these etched sheets, it was easy to align them on a microscope stage and to make the measurements by successively focusing down through the layers. A histogram of the average of four minor axes for each particle track is shown in figure 8. The charge resolution σ_z is 0.12 e. Because their surfaces were rough after etching, we could not make measurements through the layers of undoped CR-39. However, measurements of minor axes of the etch pits on the top surface only, show that the charge resolution is similar for the doped and undoped polymer.

$$\sigma_z \approx 0.23\ e/\sqrt{n}$$

where n is the number of etch pits measured per event. Practically speaking, use of the additive enables one to attain far higher resolution by virtue of being able to make etch pit measurements quickly in many successive sheets.

In experiments similar to ours, Gary D. Westfall and his colleagues[22] obtained a charge resolution $\sigma_z = 0.12\ e$ for a 3-mm-thick silicon detector without matter in front of it and $\sigma_z = 0.17\ e$ for an identical detector with matter in front of it. From the data used to make figure 8, we can say that our measurement of a particle's charge using a CR-39(DOP) detector stack with thickness equivalent to that of a 3 mm silicon detector has a standard deviation of 0.022 e, which is about six times better than the measured value for silicon. Impressive as this may sound, it falls short of the theoretical resolution given by present models of the mechanisms of track formation. Fluctuations due to residual non-uniformities in the plastic probably dominate over energy-loss fluctuations. If they can be further reduced, even high charge resolution should be possible.

We found, when taking the data in figure 8, that the locations, magnitudes, and cross sections of charge-decreasing fragmentation reactions could be determined quickly and accurately by noting the sudden decrease in the size of the etch pit mouths. By bombarding an extremely thick (about one thousand sheets) stack with high-energy iron nuclei (about 2 GeV/amu) it will be possible to explore more quantitatively the recent discovery[23] that relativistic heavy nuclei, immediately after nuclear interactions, tend to have anomalously high cross sections for further interactions. The ability to obtain greatly improved statistics and charge resolution, in comparison with the previously used nuclear emulsion, should permit added insight into the nature of the anomalous nuclei.

In the future, work at the frontier of cosmic ray astrophysics will involve accurate measurements of isotopic and elemental abundances of rare heavy elements from nickel up to uranium. This will require both large collecting power and superb resolution. Doped CR-39 is the first detector to satisfy both of these requirements, and large experiments are planned for a shuttle-borne Spacelab mission and for a two-year free-flying platform.

Searches for primary antimatter in cosmic rays have been limited by size and weight restrictions on magnets flown as balloon payloads. Such experiments have placed limits of about 1 part in 10^4 on the fraction of antimatter. It would be very interesting to be able to detect 1 part in 10^6, because it is possible that more than this fraction of cosmic rays come from other galaxies, some of which might be made of antimatter. We can attain such a sensitivity by exploiting the difference in the response of track detectors and plastic scintillators stacked in an alternating array of large area. The correct treatment of energy transfer from a charged particle to electrons in a medium involves the Mott cross section for electron scattering, which for small energy transfers is independent of the sign of the nuclear charge but for large energy transfers is larger for a positive nucleus than for an antinucleus. Because low-energy electrons produce most of the radiation damage within about 4 nm of a particle's trajectory, a track detector measures only the magnitude of the charge. In a plastic scintillator little light is emitted from the saturated core; most of the light results from energy sparsely deposited at large radial distances by high-energy electrons whose production rate is higher for a nucleus than for an antinucleus. The sensitivity of this type of experiment increases with $|Z|$ and depends crucially on the charge resolution of the detec-

tors. With a charge resolution at iron of 0.1 e for a thin sheet of CR-39(DOP), the probability of charge misassignment can be reduced to a negligible level while keeping the nuclear interaction background small.

With a 5-mm-thick CR-39 detector, we can have a standard deviation as small as 0.02 e in our measurement of charges. This would enable us to resolve any energetic fractionally charged nuclei from those with integral charges. Searches for energetic quarks bound to heavy normal nuclei are therefore possible in experiments with accelerators or cosmic rays.

Our group is doing major experiments involving CR-39 track-recorders at the Stanford–LBL positron–electron colliding-beam facility (PEP), where magnetic monopoles and other highly ionizing particles are being sought among the products of e^+ and e^- collisions, and at the tops of high mountains, where large arrays of the detectors are deployed in searches for exotic, highly charged particles in nature.

Future

History teaches us that every new detector evolves from a discovery phase, where it shows promise but is insensitive and unreliable, to a development phase, where heroic efforts, stimulated by scientific results achieved with the detector in its early form, lead to a precision instrument of great sensitivity and resolving power, and then to a steady-state phase, where the detector in its ultimate reliable form is readily available and routinely used at the forefront of research. Nuclear emulsion, the bubble chamber and the semiconductor diode detector are examples that spring to mind.

The first nuclear track-recording solid, natural mica, was extremely insensitive; it had at first to be studied by transmission electron microscopy, and it suffered from a crippling difficulty: the very electron beam used to see the tracks erased them within a few seconds! In the development phase the chemical etching technique was discovered, the track-recording property was found to be a feature of all dielectric solids, the quantitative relationship between track-etch rate and radiation-damage rate was discovered, and the uniquely high charge resolution of certain plastic track detectors was demonstrated. Even though the development phase shows no sign of drawing to an end, we believe it will soon be possible to obtain plastic track detectors with standardized response and resolution for the identification of nuclear particles. We have begun investigating polymers that may be much more sensitive to radiation than even allyl diglycol polycarbonate, so that the advantages of track-recording solids may someday extend to particles with Z/β as low as 1 or 2, and allow a concomitant expansion of scientific and technological applications.

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