Identification of Isotopes of Energetic Particles with Dielectric Track Detectors

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This paper describes a new principle of charged-particle identification by plastic track detectors. We find that the rate of chemical etching (by a suitable reagent) along the track of a heavy ion in a plastic depends only on the primary ionization rate, which, as the particle slows down, increases at a rate that depends uniquely on atomic number Z and mass A. To identify the Z, A, and energy of a particle we can either measure the etching rate at a known residual range [by analogy with a $(dE/dx)-E$ detector] or measure the etching rate at two known points along the trajectory in a stack of plastics. We present experimental data for B^{10}, B^{11}, C^{12}, N^{14}, and O^{16} ions from the Yale accelerator, showing that masses differing by 9% can be resolved. Unique features of this method are its discrimination against intense background radiation and its combination of high resolution with large collecting area, which make it feasible to do certain cosmic-ray experiments, such as the detection of Be^{10} nuclei.

DIELECTRIC detectors,[1] in which nuclear particle tracks are made visible by a preferential chemical etch, are uniquely useful for certain studies in nuclear physics, geophysics, and astrophysics. One of their most valuable features is the existence of a critical primary ionization rate such that only particles with primary ionization rates above the critical rate will produce etchable tracks.[2] This property has been exploited as a means of setting lower limits on the masses of those particles that record tracks. Inorganic crystals have been used, for example, to measure formation cross sections and angular distributions of ternary fission fragments without any background from an impinging beam of argon ions,[3] and to measure the abundance of cosmic rays with $Z>30$, relative to the abundance of iron nuclei.[4,5]

In this paper we show how dielectric detectors can be used to identify atomic numbers and to distinguish between isotopes of certain energetic nuclei. Although the data reported here apply only to one brand of cellulose nitrate, the principle is valid for other plastics and possibly for other dielectric solids. This discovery extends the usefulness of dielectric detectors and makes feasible the identification of certain energetic isotopes in solar and cosmic rays and of light nuclei emitted in asymmetric fission. The principle of charge resolution is based on our observation, to be described below, that the rate of chemical etching along a particle track in a plastic depends only on the primary ionization, which is a function of the velocity and the atomic number Z of the particle. Thus the identification of Z for a given particle requires only a simple measurement of etching rate at two known points on its trajectory in a stack of plastic sheets. For nuclei having the same charge, the residual ranges for a given velocity are proportional to their masses. Hence the etching rate along their paths expressed as a function of residual range will be different for each mass number. In principle, therefore, it is feasible to distinguish between the isotopes of certain nuclei by measuring the etching rate (in practice the etched track length) at a point on the trajectory of each isotope, and its residual range from that point.

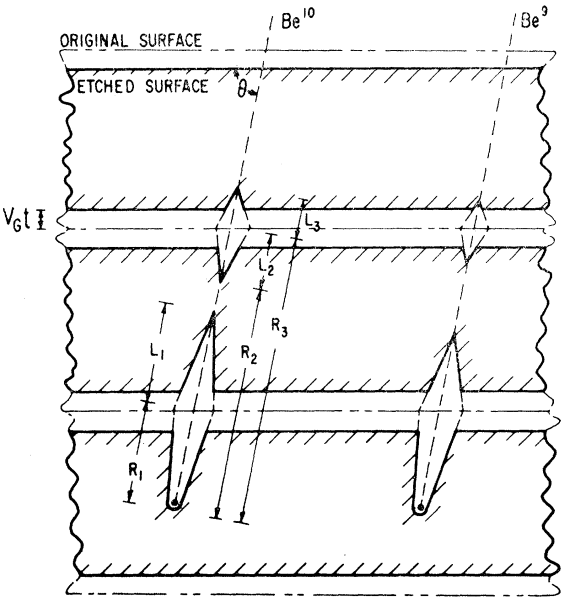

FIG. 1. Principle of particle identification by dielectric detectors. In a hypothetical situation where ions of Be^{10} and Be^{9} penetrated to the same depth in a plastic stack, the rate of etching of the cone-shaped tracks L_i is faster for Be^{10} than for Be^{9}.

[1] For a review of the properties of dielectric track detectors, see R. L. Fleischer, P. B. Price, and R. M. Walker, Ann. Rev. Nucl. Sci. **15**, 1 (1965).
[2] R. L. Fleischer, P. B. Price, R. M. Walker, and E. L. Hubbard, Phys. Rev. **156**, 353 (1967).
[3] R. L. Fleischer, P. B. Price, R. M. Walker, and E. I. Hubbard, Phys. Rev. **143**, 943 (1966).
[4] R. L. Fleischer, P. B. Price, R. M. Walker, M. Maurette, and G. Morgan, J. Geophys. Res. **72**, 355 (1967).
[5] P. B. Price, R. S. Rajan, and A. S. Tamhane (to be published).

In the Yale heavy-ion accelerator, we exposed stacks of 250-μ-thick sheets of clear Nixon-Baldwin cellulose nitrate to 10.3 MeV/N ions of B^{10}, B^{11}, C^{12}, N^{14}, and O^{16}, using wedge-shaped absorbers to produce a spectrum of energies from 0 to 10.3 MeV/N. At Schenectady and Dublin we etched stacks for 9 h at 20°C in stirred solutions of 6.25N NaOH and measured the features, shown in Fig. 1, that can be used to identify the particles. After an etching time t, a thickness $v_G t$ is removed from the surfaces of each sheet, where v_G is the rate at which the plastic is dissolved away. Cone-shaped "tracks" L_1, L_2, L_3, $\cdots$ are etched into the plastic wherever the primary ionization rate J of a particle crossing a surface exceeded the critical rate J_c. The observed lengths are directly measured; the *true* lengths are calculated by correcting the observed lengths for the portions $\Delta L = v_G t \csc\theta$ that were lost in the etching process. The true lengths L_i provide a measure of the ionization rate along the particle trajectory:

$$L_i = \int_0^t v_T(i)dt = \int_0^t f(J - J_c)dt,$$

where f is a function to be determined below and L_i is defined only for $v_T > v_G$. Thus v_T is not constant along the trajectory but increases with ionization rate and is highest near the end of the range of a particle.

For each measured track length L_i, there can be measured a residual range R_i, defined as the distance from the low-energy end of the segment of length L_i to the point where the particle comes to rest in the stack. After all measurements of L_i have been made, the sheets are reetched for a time sufficient to ensure that the reagent has penetrated to the ends of the ranges of the particles, which leads to the formation of tracks having characteristic rounded tips. We have found that ranges of ions of known energy deduced from measurements of tracks with rounded tips agree within 2% with ranges calculated from the range-energy curves of Henke and Benton.[6]

Measurements of L_i for B^{10}, B^{11}, C^{12}, N^{14}, and O^{16} ions in different sheets of cellulose nitrate are plotted as a function of R_i in Fig. 2. At the maximum energy of 10.3 MeV/N, all ions penetrate one 250-μ sheet and part of a second 250-μ sheet. For both the Schenectady and Dublin data there is a clear tendency for tracks of B^{11} ions to be longer than tracks of B^{10} ions with the same residual range. The straight lines through the B^{11} and B^{10} data, calculated by the method of least squares, intersect the abscissas at values of residual range having ratios ~11/10, as is to be expected.[7]

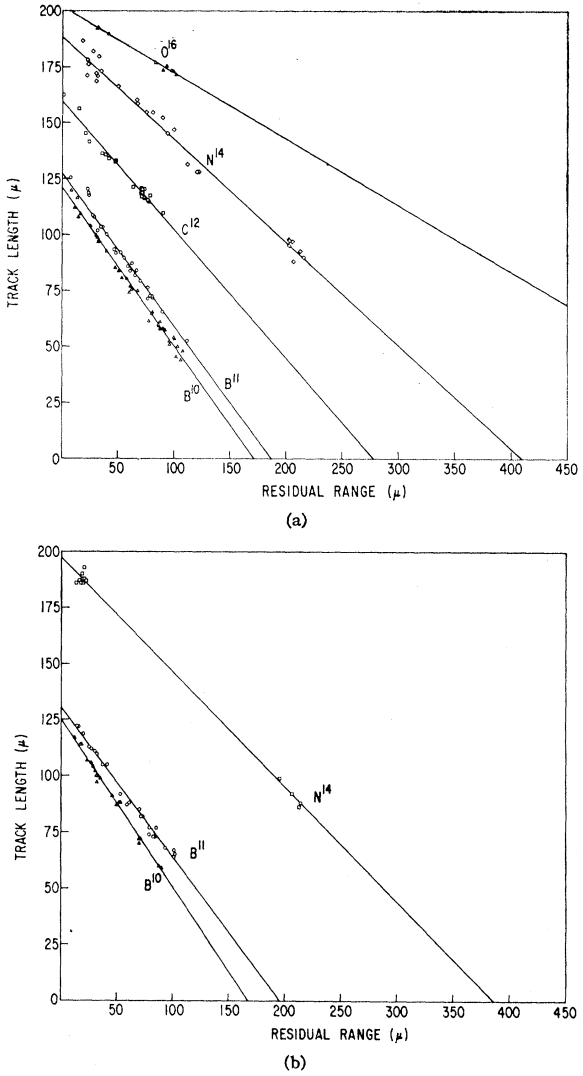

FIG. 2. Measurements of track length as a function of residual range for B^{10}, B^{11}, C^{12}, N^{14}, and O^{16} ions in different stacks of cellulose nitrate: (a) etched 9 h and examined at Schenectady; (b) etched 9 h and examined at Dublin. Each experimental point applies to a different track.

Although the reproducibility of etching rates over large areas and in many nominally identical sheets of cellulose nitrate remains to be established, these results indicate that in small, calibrated sheets it is already possible on a statistical basis to distinguish between isotopes of energetic ions differing in mass by about 9%, simply by measuring the two parameters L_1 and R_1. Additional measurements L_2, R_2, L_3, R_3, etc., would provide further information and may make it possible to distinguish isotopes with a smaller mass difference.

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

[7] Any differences between the positions of the least-squares lines fitting the Schenectady and Dublin measurements which do not arise from statistical uncertainties may arise from small differences in the conditions of etching and in microscope optics. It is intended, in the future, to refine further the conditions of etching and to protect the samples from the effects of humidity, which may have the effect of obscuring the tips of the tracks, especially at Dublin where the ambient humidity is commonly higher than at Schenectady. At Schenectady we have investigated the effect of rinsing the samples in ultrasonically agitated hot distilled water. It has been found that this process improves to a marked degree the visibility of the tips.

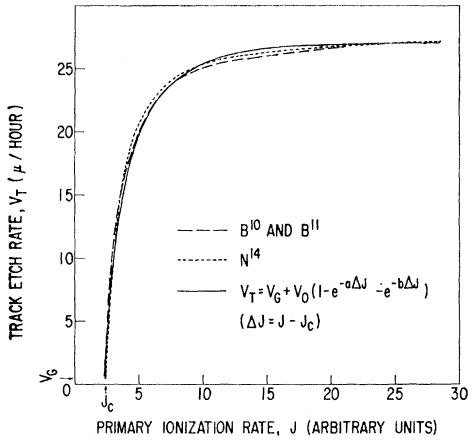

FIG. 3. Dependence of etching rate on primary ionization rate, calculated from the data as described in the text. From the shape of this curve it is clear that the best resolution of ions of similar mass is obtained when the ionization rate is not far above threshold, J_c. At high ionization rate the etching rate approaches a constant maximum value of about 27 μ/h.

By differentiating the least-squares lines of Fig. 2(a) with respect to time, we have obtained etching rates as a function of residual range. Then, using curves of relative primary ionization rate versus energy[2,8] together with range-energy curves,[6] we have deduced etching rates as a function of relative primary ionization rate, as shown in Fig. 3. The important conclusion to be drawn from Fig. 3 is that etching rate increases with ionization rate in the same way for each isotope.

We have found that the relationship

$$v_T = v_G + v_0(1 - e^{-a_1\Delta J} - e^{-a_2\Delta J})$$

(where $\Delta J = J - J_c$) is a useful empirical fit to the points in Fig. 3 and can be used to calculate curves of L versus R for isotopes like Be^{10} that are not at present available in heavy-ion accelerators. This equation, based on first-order chemical kinetics, assumes that the increase in chemical reactivity along a track is due to irradiation-produced changes in the relative concentration of chemically etchable species.[9]

It now appears feasible with rocket, balloon, or satellite exposures of plastic stacks to estimate the isotopic composition of Li, Be, and B ions in solar and cosmic rays, provided the abundances of adjacent isotopes are not too disparate. At present this does not appear to be possible either with semiconductor detectors (because of the low counting rates) or with nuclear emulsions. Such measurements would have a number of important astrophysical applications. For example, among the isotopes produced in interstellar spallation reactions is Be^{10}, which, because of its long half-life (2.7×10^6 years), can be used as an indicator of the age of the cosmic rays and of the density of the region in which most of their time was spent.[10] Information about the composition of cosmic-ray sources and about the solar atmosphere could be gained from measurements of the isotopic composition of C, N, and O nuclei in cosmic and solar particles.

In another paper we will show that the etching rate of Lexan polycarbonate, which has a higher critical ionization rate than cellulose nitrate, does not saturate even when bombarded with ions as heavy as zinc. It should therefore be useful in a mass region well above that for which cellulose nitrate is best suited. We estimate that relativistic cosmic rays with atomic number in the vicinity of 90 [11] could be identified with an uncertainty of only 3 to 4 units.

Note added in proof. In a current study of a large stack of Lexan sheets that were exposed to cosmic rays at Fort Churchill, we have been able to resolve all the elements from neon ($Z=10$) up to at least zinc ($Z=30$). The etching rate along a track in Lexan is found to increase exponentially with primary ionization, so that the resolution of adjacent elements actually *improves* with increasing Z. Assuming this same dependence holds for relativistic particles, we calculate that tracks of plutonium nuclei would be $\sim$20% longer than tracks of uranium nuclei and thus easily distinguishable.

A significant advantage of plastic detectors in nuclear physics is their ability to record tracks in the presence of a strong flux of electrons, γ rays, neutrons, and high-energy protons and α particles, none of which register in the detector. One could, for example, use thin plastic sheets together with mica to measure the masses of very light, long-range fission fragments[12–14] and to discover whether they are formed in asymmetric binary fission or in ternary fission. One could also identify long-range heavy nuclei produced in high-energy spallation reactions.

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[8] Although absolute values of primary ionization in plastics are not known, we have shown in Ref. 2 that relative values obtained from the modified Bethe equation allow one to explain track registration data in plastics. The units of primary ionization rate in Fig. 3 are the same as those in Ref. 2.

[9] A detailed description of the etching-rate equation will be postponed until more specific evidence of the nature of the reactions involved can be produced.

[10] B. Peters, Pontificiae Academiae Scientarum Scripta Varia **25**, 1 (1963).

[11] P. H. Fowler, R. A. Adams, V. G. Cowen, and J. M. Kidd, Proc. Roy. Soc. (London) **A301**, 39 (1967).

[12] M. L. Muga, in *Proceedings of the Symposium on the Physics and Chemistry of Fission* (International Atomic Energy Agency, Vienna, 1965), Vol. II, p. 409.

[13] R. H. Iyer and J. W. Cobble, Phys. Rev. Letters **17**, 541 (1966).

[14] J. Chwaszczewska, M. Dakowski, T. Krogulski, E. Piasecki, W. Przyborski, and M. Sowinski, Phys. Letters **24B**, 87 (1967).