CERN-EP/84-72
June 22nd, 1984

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We describe how to produce sheets of CR-73, a polymer similar to bisphenol-A polycarbonate in its monomer unit but with a cross-linked structure similar to that of CR-39, a diallyl polycarbonate commonly used as a track-recording plastic. We have analyzed etched tracks of 0.95A GeV U ions, of 1.45A GeV Kr ions, and of projectile fragments of 1.2A GeV La ions in CR-73. In the charge region $45 \leq Z \leq 57$ the charge resolution for a single etch pit, $\sigma_Z \approx 0.18e$, is significantly better than that of the commonly used detectors Tuffak polycarbonate ($\sigma_Z \approx 0.29e$) and CR-39 ($\sigma_Z \approx 0.5e$). The minimum detectable value of Z/β for normally incident particles is ~ 40 for CR-73 compared with ~ 52 for Tuffak.

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1. Introduction

With the availability of beams of relativistic nuclei up to uranium at the Bevalac, it has become possible to make accurate measurements of the response and charge resolution of solid-state nuclear track detectors [1-4]. Two such detectors stand out as applicable to research in a variety of fields including cosmic ray astrophysics, relativistic heavy ion physics, and searches for hypothetical particles such as supermassive magnetic monopoles. One of them, CR-39(DOP), a diallyl polycarbonate doped with 1% dioctyl phthalate [5], has been shown [2] to have a charge resolution $\sigma_Z = n^{-\frac{1}{2}} (0.12 + 0.006 Z^*/\beta) e$ in the interval $10 \leq Z^*/\beta \leq 90$, where n is the number of etch pits whose major axes are measured in a stack of thin sheets and Z^* and βc are the effective charge and velocity. (For a description of track-etch detectors and techniques, see [6].) The other, Tuffak polycarbonate, has a charge resolution that ranges from $\sigma_Z \approx 0.29 n^{-\frac{1}{2}} e$ at $Z^*/\beta \approx 60$ to $\sigma_Z \approx 0.85 n^{-\frac{1}{2}} e$ at $Z^*/\beta \approx 90$ to 105. These two detectors complement each other, CR-39(DOP) being optimal at values of Z^*/β below ~ 80 and Tuffak being preferable (partly because of its low cost and availability in thin sheets) at higher values of Z^*/β .

As part of a search for materials with even better charge resolution than CR-39(DOP) and Tuffak, we report here a study of a polymer called CR-73, which we selected in the expectation that it would combine the superb optical qualities and etched track shapes characteristic of CR-39 with the lesser sensitivity characteristic of Tuffak. (There are applications where it is advantageous to be able to record only particles with very high Z^*/β .) Our reasoning stems from an examination of the chemical structure of CR-39, Tuffak and CR-73 shown in Fig. 1.

CR-39 is a large polycarbonate molecule with diallyl functionality. Polymerization of the monomer can proceed from both ends via free-radical initiation. The resulting polymer is a thermosetting, cross-linked network that is insoluble and highly uniform. Radiation damage is known to occur at relatively low doses of radiation [7], there being no aromatic rings to soak up energy nondestructively. Etching proceeds, as in all polycarbonates, by hydrolysis of the carbonate linkage, making it susceptible to chemical attack by strong bases and acids. Unlike CR-39 polymer, Tuffak polycarbonate is produced from a polymer melt by extrusion. During cooling below the glass transition temperature, crystalline regions can be formed, resulting in an inhomogeneous polymer. We suspect that Tuffak has a higher uniformity than Lexan (also made from bisphenol-A polycarbonate) because it is allowed to relax just after extrusion and before rolling. Both Lexan and Tuffak are more resistant to radiation damage than is CR-39, undoubtedly due to the energy-absorbing properties of the aromatic rings (Fig. 1). CR-73 has the diallyl functionality of CR-39 with the aromatic ring structure of Tuffak. CR-73 is cast from the monomer, just as is CR-39, into a hard, thermosetting, cross-linked polymer. It thus should have the high uniformity of CR-39 and yet have the low radiation sensitivity of Tuffak.

2. Experimental Results

To test our hypothesis we obtained a sample of CR-73 monomer from PPG Industries and polymerized it according to the same procedure used for CR-39. We first added 2.5% isopropyl peroxydicarbonate (IPP) and then introduced it into a mold made from two pieces of float glass separated by an Elvax gasket. The

entire assembly was heated in an air oven with a temperature cycle beginning at 40°C and rising to 100°C in 40 hours. The resulting polymer was an optically clear, brittle plastic with a specific gravity of 1.22 and a refractive index of 1.56.

Using the Lawrence Berkeley Laboratory Bevalac, we did three sets of irradiations. We exposed a stack of 0.8 mm-thick CR-73 sheets at normal incidence to 1.2A GeV ^{139}La ions, using a 2.54 cm slab of polyethylene to produce projectile fragments with essentially the same velocity as that of the La beam; we exposed other sheets of CR-73 to 1.45A GeV Kr ions at normal incidence and to 0.95A GeV U ions both at normal incidence and at a zenith angle of 34° . We etched the CR-73 sheets 94 hours at 70°C in 6.25 normal NaOH solution and measured the thickness of material removed from each surface. Both from direct measurement of thickness before and after etching and from measurements of etched track diameters and lengths, we obtained the value $v_G t = 70\text{ }\mu\text{m}$, where v_G is the general etch rate.

a) La results

We superimposed one CR-73 sheet from a depth 2.9 g cm^{-2} in the stack on the first sheet, aligned the two sheets by means of the pattern of beam tracks, focussed on the interface between the two sheets, and scanned for beam tracks whose diameters indicated that they had been produced by La ions with the correct beam velocity. We rejected about 3% of the tracks with the wrong diameter, which resulted from fragmentation or slowing in upstream matter, mainly the walls of the beam pipe.

We used a Leitz Ortholux microscope with a 45X dry objective, an eyepiece filar micrometer, and transmitted light to measure etch pit diameters. Figure 2 shows our results. The

shaded histogram is the distribution of etch pit diameters obtained in an unbiased scan. The unshaded histogram includes additional events that did not lose charge in passing through the polyethylene.

The unfragmented La beam ($Z = 57$) is cleanly resolved from the adjacent peak corresponding to fragments with $\Delta Z = 1$, and individual charge peaks are identifiable by eye down to $Z \approx 46$. Figure 3 is a plot of the positions of peaks in the histograms as a function of charge, on the assumption that they define a smooth curve. The arrows in Fig. 2 indicate the expected positions of charge peaks if the curve is a straight line determined by a fit in the interval $46 \leq Z \leq 57$.

Knowing the distance between charge peaks, we determined the charge standard deviation for a single etch pit measurement. A gaussian fit to the peak corresponding to the unfragmented La beam gives $\sigma_Z = 0.18e$. For charges 46 through 56 we shifted all measured track diameters by the appropriate integer multiple of the average separation between charge peaks, so that the individual peaks coincided. From the full width at half maximum of the resulting histogram, we obtained the value $\sigma_Z = 0.17e$. In the same charge interval the charge standard deviation for Tuffak is $0.29e$ [3]. For CR-39(DOP), $\sigma_Z \approx 0.5e$ [2].

The sensitivity, defined as $s = v_T / v_G$, where v_T is the etch rate along the track, is related to etchpit diameter by the expression

$$s = (1 + D^2 / (2v_G t)^2) / (1 - D^2 / (2v_G t)^2).$$

Figure 4 compares our results for CR-73 with the responses of Tuffak and CR-39(DOP).

b) Kr results

Etchpits due to 1.45A GeV Kr ions are barely detectable at normal incidence. The value of reduced track etch rate resulting from our diameter measurements is plotted in Fig. 4.

c) U results

Using the eyepiece micrometer to measure diameters of the normally incident tracks, we obtained a fractional standard deviation of 0.006, which leads, through the relation

$$\sigma_D/D = \sigma_s/(s^2 - 1)$$

to a fractional standard deviation $\sigma_s/(s-1) = 0.06$. From the slope of the response curve in Fig. 4, we compute from this result a charge standard deviation $\sigma_Z/Z = 1.0e$, a value only slightly poorer than was obtained earlier [1,3] using a Compumetric video system to measure etchpit diameters of U ions in Tuffak.

Using a semi-automated track profile method [8] to measure lengths of the tracks at a 34° zenith angle, we obtained a mean value $s = 9.43$ with a fractional standard deviation of 0.035 and a single-event reproducibility of 0.003. (Using a 22X oil objective and the eyepiece micrometer to measure track lengths we obtained a mean value $s = 9.32$.) The corresponding single-etchpit charge resolution, $\sigma_Z/Z = 0.7\%$, is better than we were able to obtain using the eyepiece micrometer to measure diameters and is close to the limiting resolution set by electron capture and loss processes [1]. It is comparable to the resolution of Tuffak for U ions [3].

3. Discussion

Some years ago B. G. Cartwright predicted [9] that cross-linked, thermoset polymers would be superior to the thermoplastic

solvent-cast polymers then being used as track detectors. He also pointed out that track-etch detectors should in principle have charge resolution superior to that of any other detector used in a transmission mode. The reason is that track-etch detectors are sensitive mainly to very low-energy transfers, for which the fluctuations that limit detector resolution are very small compared with fluctuations in high-energy transfers, which contribute importantly in other types of detectors. Cartwright and co-workers [9] reported that CR-39, a cross-linked thermoset polymer, showed promise of having remarkably high charge resolution.

In the present study we have confirmed Cartwright's predictions by providing a second example of a cross-linked thermoset polymer, CR-73, with remarkably high charge resolution. Using the restricted energy loss model of track formation, in which only energy transfers less than w_0 contribute to track formation, Ahlen [10] has obtained the following expression for the standard deviation of the average value E of the restricted energy loss of a particle of charge Z and velocity β in passing through a thickness x of a detector:

$$\frac{\sigma_E}{\Delta E} \approx \frac{0.0015 [w_0(\text{eV}) + 2640 + 656 \ln \beta]^{1/2}}{Z [x(\text{cm})]^{1/2} (4.06 + \ln(\beta\gamma))} \quad (1)$$

For a 1.2A GeV ^{139}La ion, a value of $w_0 = 350$ eV [2], and an effective thickness of detector material given by $x = v_G t s / (s+1) = 0.004$ cm, we expect on the basis of (1) that $\sigma_E / \Delta E \approx 0.004$. As discussed in [2], we can compute the corresponding charge standard deviation due to fluctuations in restricted energy loss. The result is that $\sigma_E / \Delta E = 0.004$ corresponds to $\sigma_Z = 0.12e$ for 1.2A GeV La ions. We conclude that the resolution of CR-73 is

close to the theoretical limit imposed by fluctuations in energy transfer to constituents (mostly electrons) of the detector. For an equivalent thickness, $\sigma_E / \Delta E$ is about 20 times larger for a total energy loss detector such as a semiconductor detector.

From Fig. 4 one sees that the sensitivities of CR-73 and Tuffak are comparable at high ionization rate but that $s-1$ falls less steeply with decreasing Z^*/β for CR-73 than for Tuffak, so that it can be used over a wider charge interval. If CR-73 can be produced commercially at a reasonable price, it will be preferable to Tuffak polycarbonate when the ultimate in resolution for $Z^*/\beta \gtrsim 50$ is demanded.

This work was supported in part by NASA Grant NGR 05-003-376. We thank Michael Solarz for valuable assistance. P. B. Price is indebted to CERN for a Scientific Associateship and to G. Vanderhaeghe for the loan of microscope equipment.

Figure Captions

1. Structure of CR-39 monomer (diethylene glycol bisallyl carbonate), Tuffak polymer (bisphenol-A polycarbonate), and CR-73 monomer (bisphenol-A, bisallyl carbonate).
2. Distribution of etch pit diameters for 1.2A GeV ¹³⁹La and its projectile fragments detected in a single sheet of CR-73. Shaded histogram is for an unbiased scan; unshaded histogram is for a scan for fragments with $\Delta Z \geq 1$.
3. Diameter as a function of charge obtained from peaks in the histogram in Fig. 2.
4. Reduced track etch rate as a function of Z^*/β for CR-73, Tuffak polycarbonate, and CR-39(DOP).

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Fig. 1

Fig. 2

Fig. 3

Fig.4