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DEVELOPMENT OF PASSIVE RADIATION DETECTORS OF IMPROVED SENSITIVITY

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ABSTRACT

In this paper we discuss the future development of a solid track high energy particle detector. Our goal is to improve the sensitivity and lower the threshold of the detector. One most widely used material for such purpose is a plastic commercially known as CR-39. We present a scheme which involves in changing the formula of the monomer, diethylene glycol-bis-allyl carbonate. This is to be accomplished by substituting some heteroatoms for H and substituting sulfur atoms for oxygen in the ether linkages.

We also suggest use of a new plasticizer to make the etched surface clearer than what has been accomplished as of today.

Possible improvement in acquiring better tracks and increasing the ratio of V_T/V_B has been planned. This is to be accomplished by changing the composition of the etchants, etching time, and etching temperature.

OBJECTIVES

1. Survey the materials and applications of etchable nuclear particle track detector field.

2. Determine the cosmic ray detector materials most likely to be adaptable to controlled changes in sensitivity.

3. Recommend approaches to controlled sensitivity changes which will lower the threshold detection of the etchable detectors in terms of the primary particle's charge (Z) and its velocity parameter ($\beta^{-1} \equiv \frac{c}{v}$).

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INTRODUCTION

The first published observations of individual radiation damage tracks in solids were by Young (1958) /1/ and Silk and Barnes (1959) /2/ who reported etched U235 fission fragment tracks in LiF and transmission electron micrographs in mica, respectively. Stimulated by the Silk and Barnes observations, and other scientific and practical problems at the time, R. L. Fliescher, P. B. Price, and R. M. Walker investigated the revealation of the radiation damage tracks by strong etching baths for crystalline solids, amorphous solids (glasses), and plastics (artificial glasses). As the applications rapidly expanded: controlled micro-vacuum leaks, filters, archeology, geochronology, cosmogeny, cosmic ray physics, particle physics, neutron and heavy particle dosimeter, etc.; many investigators joined the field, either to further development of the technique or to apply it to their particular speciality.

The development of the technique and application to various fields are comprehensively described in "Nuclear Tracks in Solids" by Fliescher, Price, and Walker (1975) /3/. Their detailed literature search (described in their book which contains 1500 references) ensures that this is the correct starting point for a study of etchable particle track detectors. Since 1975, other materials, techniques, and applications have appeared. A good survey of the recent development for to cosmic rays and particle physics can be performed by screening articles in Nuclear Tracks and Radiation Measurements /4/ (published quarterly since 1980), Nuclear Instruments and Methods /5/, and the biennial "Papers of the XX Cosmic Ray Conference." /6/ Other journals, conference proceedings, contract reports, etc., may, of course, contain important information, particularly about applications in dosimetry, geology, etc.

The three general classifications of etchable track detectors are crystalline solids, amorphous solids (generally glasses) and polymer plastics (occasionally called artificial glasses). Some naturally occurring material used in geo or lunar chronology may combine the characteristics of the first two classifications. An abreviated survey of detector materials is given in Table 1.

The ability to preferentially etch the radiation damage tracks depends upon the modification of the material by the primary particle to a sufficient extent that strong etching

Table 1. Some Typical Particle Track Materials

Minerals and Crystals	- beryl, calcite, feldspar, garnet, gypsum, mica, alivine, quatz, topay, zircon
Glasses	- alumina-silicate, basaltic, borate, flint, obsidian, fused quartz and fused silica, soda-lime, tektite
Plastics	- cellulose acetate, cellulose nitrate, polyrinide, polycarbonate (e.g., Lexan), polymethyl methacralate (Plexiglass), polystyrene

solutions may etch the material at a higher rate along the radiation damage path than at the surface of the bulk of the material. Therefore, the track etch rate (V_T) and the undamaged or bulk etch rate (V_B) are the two parameters that determine the detector behavior.

V_T depends upon the radiation damage to the detector material. In crystalline materials the damage is obviously crystalline disorder. In glasses it must be changes in the amorphous order-disorder condition, or locally where damaged a "different kind of glass." For organic polymer materials the principal mechanisms of damage must be the breaking of chemical bonds. Both direct bond breaking by ionization and some induced atomic disordering may be involved.

The energy transfer process from the primary particle to the detector material that is damaged has been the subject of much study. In addition to the references mentioned above, the "Werner Brandt" Conference Proceedings /7/ carry much on this subject. At present only a general schematic of the entire energy transfer to radiation damage process is available. The net result is crystalline lattice damage, amorphous glass structure changes, and broken polymer chains in the three material classifications, respectively. Detailed mechanisms such as the "ion explosion spike" in crystalline materials, the "primary ionization" or "restricted energy loss" in polymer materials are still debated. It is clear that the total ionization energy loss of the primary particle as described by the Bethe Bloch formula does not correlate well with the etched detector response. /8/

The ionization energy loss of the primary particle is partially transferred to many individual electrons in the material, many of which are given significant energies and travel some distance from the particle track. That these electrons are not effective in providing track damage is borne out by the apparently small radius of the damage region (generally less than a micrometer). The small size of the damage region makes radiation damage mechanisms difficult to investigate. For some polymer plastics it seems that a "restricted energy loss that neglects some, but not all energy transfers to electrons, fits the response reasonably well. /9/ Therefore, even yet, each detector type, and sometimes each detector batch, must be subjected to calibration if used over a wide range of primary particle velocity (v) and primary particle charge (Z). The etchable track detectors generally have a rather high "threshold" of primary particle charge (Z) and inverse particle velocity (β^{-1}). Some crystalline and glass detectors only respond to slow fission fragments or recoil nuclei. The presently most

sensitive reliable detectors are organic polymers (CR39 plastics) which have thresholds at relativistic oxygen nuclei (Z = 8) at normal incidence to the detector surface. That the threshold is incident angle dependent is shown in Figure 1 which demonstrates the effect of the bulk etch rate (V_B) in reducing the effectiveness of the track etch rate (V_t) at incident angles off the zenith. For many applications, such as the one of concerned here, the particles are mostly not incident normal to the plastic surface. In fact, the primary particle will be typically inclined at ~45° to the surface, raising the threshold to Z > 20.

There are many applications for etchable detectors of greater (or lesser) sensitivity. The one related to this work requires a greater sensitivity and this requirement will be briefly described here. It is the use of etchable plastic sheets in emulsion chamber experiments of the type flown by the JACEE collaboration. /11/

The use of the track detectors in the JACEE emulsion chamber is for determining the primary cosmic ray charge as it enters the emulsion chamber and the heavy fragment(s) of the primary after any interaction which may occur in the chamber (which occur with ~20% probability with cosmic ray protons and ~90% probability with cosmic ray iron nuclei). The nuclear track emulsions can also supply the charge information to z ± 2, but the etchable plastic can be accurate to Z ± 0.5. The higher accuracy is very important to the cosmic ray astrophysics objectives and to the nuclear interaction objectives of these experiments. The most sensitive etchable plastic that is reliably available is CR39, which has a threshold too high to see all the primaries at large inclinations, /11/ and registers almost none of the interaction products. An improvement in sensitivity would be very desirable. Although there are many materials in use for etchable plastic detectors today CR39 is almost the universal choice for high sensitivty etchable detectors for large area applications. Its commercial availability, uniform qualities, and low price are a result of its use in the plastic corrective lens industry. Its chemical suitability to track detector work is accidental. Nevertheless, its suitability for this work is indicated by the requirements for a good particle detector listed in Table 2.

The following factors may be considered to improve the sensitivity of polymer plastic detectors:

1. Strength of monomer bonds

Table 2. Characteristics of a Good Track Detector

1. Optically clear material, available in thin sheets ($\lesssim 10^{-3}$ m) of very uniform thickness (~1%), and large area (~1 m^2).

2. Optically smooth surface before and after etching, so that etch pits on both surfaces from the same track may be viewed.

3. Insensitive to photons and electrons in etching properties, but sensitive particle and energy of choice.

4. High ratio of V_T/V_B. V_T and V_B uniform over the entire area.

5. Relatively insensitive to detector temperature at time of track registration and the latent radiation damage track does not fade (anneal) at temperatures below ≈40 °C.

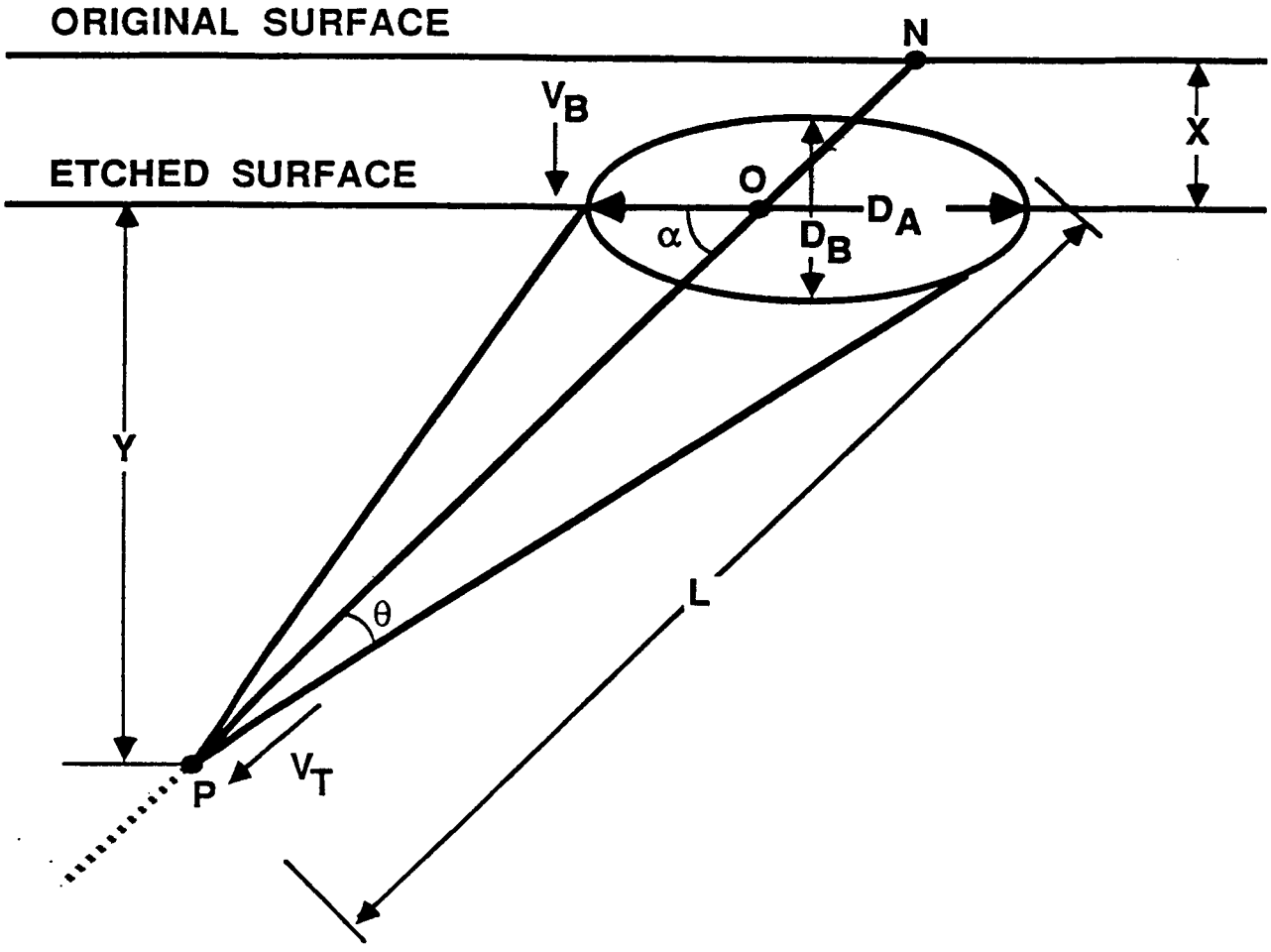


Figure 1. Geometry of typical etched cosmic ray tracks in polymer detectors. For a high energy cosmic ray ion nucleus track in CR-39, and for an etch with 7.0 normal NaOH, 70 °C, for 24 hours, X will be about 40 micrometers and D_B about 50 micrometers. X depends on the bulk etch rate V_B and etching time. The track length L (length N-P) depends on the track rate V_T, the etching time and the angle α, V_T depends on the primary charge Z, its velocity β (β ≡ v/c where c is the velocity of light) according to an empirical relationship[9] of the form $V_T/V_G \propto 1 + (Z/a\beta)^b$ where a and b are determined by calibration.

2. Polymer structure developed by plasticisers

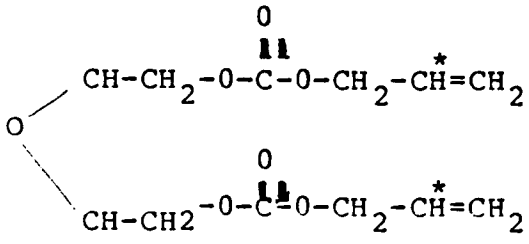
3. Polymerization process as affected by the casting and curring process

4. Effect of etching chemistry.

Factors 1, 2, and 4 will be described in the following section. Procedures of mixing and casting become more of an art. Of course the effect of any chemical modifications must be investigated experimentally also.

Chemistry and Physics of CR-39

CR-39 which is a polymer of diethylene glycol-leis-allyl carbonanate



is a thermosetting plastic. Figure 2 gives a part of the polymer. However, many other crosslinkings may exist. Its optical properties are commercially desirable. Its properties such as its transparency, mechanical stability, and sensitivity to radiation track damage are amongst the best. It is believed that damage is caused by the cleavage of some of the chemical bonds producing debris major products of which are 2, 2' oxydiethanol and polyallyl alcohol which could be swept away by some etchants. The cleavage is presumed to take place at the ester linkage. Some authors also believe that the high energy particles cause ionization by knocking out the nonbonding electrons of the oxygen atoms of the polymer. This damaged molecule is then susciptible to attack by the etchant, more so than the bulk of the material. Since our principal objective is to increase the size of the damaged zone and make the plastic more sensitive, the following suggestions are being made. If ionization is the main process responsible for the damage then a halogen such as chlorine, bromine, or iodine could be used to replace the H atoms marked by an asterisk in the above formula. The ionization potential of H and the halogens are given in the table below (L. Pauling /11/).

Figure 2. Part of the polymer CR-39. This is one of the many ways the crosslinking could take place.

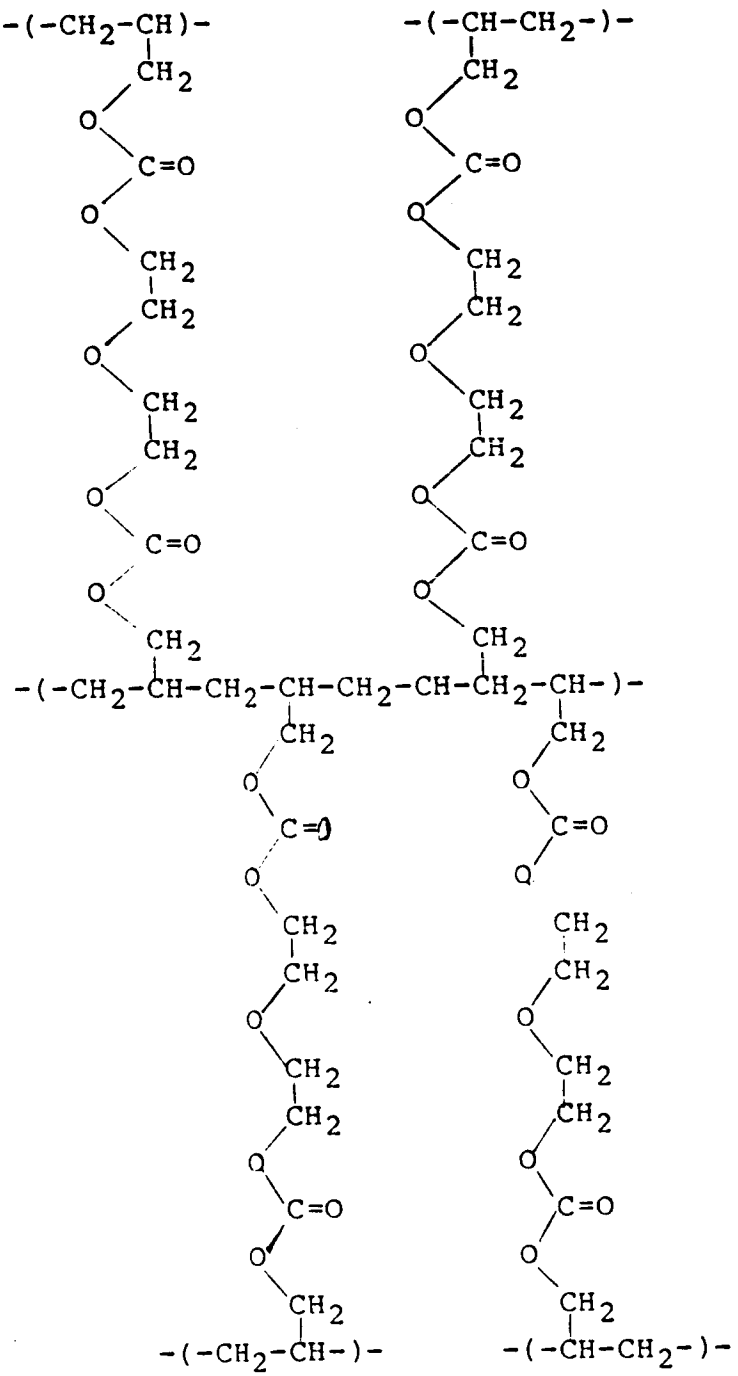


Table 1. First Ionization Potentials, I_1, of some elements

Element	I_1 Kcal/mol*
H	313.4
Cℓ	300.0
B_γ	272.0
I	241.0
S	238.8
O	313.8

*I_1 is the first ionization potential of the elements.

From the above table it appears that replacement of the H atoms by any of the halogens will increase the probability of ionization at these sites.

On the other hand if bond breaking is more desirable we still have an advantage with the substitution of the H with one of the halogens. The bond strength of C and other of the above mentioned elements are given below /11/.

Table II. Strengths of Some Bonds

Bond	Bond Strength (Kcal/mol)
C-H	98.8
C-Cℓ	78.5
C-Bγ	65.9
C-I	57.4
C-O	84.0
C-S	62.0

Here again we see that carbon-halogen bonds are weaker and easier to break than carbon-hydrogen bonds.

On the other hand, a change in the monomers may be made by replacing the oxygen of the ether group by sulfur. In Table 3 we show the ionization energies of O and S. The figures indicate that sulfur loses electrons more readily than O. Bond strength data also indicate that it is easier to break a C-S-C bond than a C-O-C bond.

Use of suitable plasticizer such as DOP (dioctylphthalate) in CR-39 plastic by Tarle /12/, showed considerable improvement, in that after doping the nonhomogeneity in the bulk of the plastic due to clustering of branched chains decreased. Nonhomogeneity in the bulk CR-39 makes the surface cloudy which introduces measurement errors and reduces the ability to see the same track on both surfaces, and to find small etch pits. We suggest that some other compounds could be tried out to yield better results in this direction. Some other authors used dinonylphthalate, DNP, in place of dioctylphthalate and obtained results similar to the ones doped with DOP.

We suggest the introduction of long hydrocarbon chains to the carboxyl and phenolic group of sallycylic acid and to use this as a plasticizer. This class of compounds are structurally similar to DOP and DNP.

Etching of the exposed samples of CR-39 is a very important factor in order to locate the tracks and determine the properties and energies of the cosmic ray particles. One of the problems encountered in etching is the ratio of V_T/V_B where V_T is the track velocity of the etching reaction and V_B is the bulk velocity. Ideally one would like to have this ratio to be infinity. Many different kinds of etchants have been tried with varying success. Most commonly used etchant is a 6.25 N NaOH solution at 70 °C. Other authors (Bean and DeSorbo, unpublished work mentioned in Fleischer et al. /3/) have tried 3.1 N NaOH solution at 7.2 °C. The etching time was 75 minutes. V_T/V_B was found to be about 10^4.

The temperature dependence of V_T and V_B had also been studied /3/ and over the range of 7 °C to 55 °C both the velocity displayed Van't Hoff dependency.

$$V_T = V_o \exp(-E_T/kT) \text{ and } V_B = V_o \exp(-E_B/kT)$$

The activation energy ratio E_B/E_T was approximately equal to 0.793, where E_T and E_B are the activation energy of the track and the bulk respectively.

Three different types of etchants should be tried:

(1) Basic type such as alkalimetal hydroxide mixed with oxidants such as $C\ell O^-$, MnO_4^-

(2) Acidic type such HNO_3, a mixture of $K_2Cr_2O_7$ and concentrated sulfuric acid, etc.

(3) Neutral type such as $KMnO_4$, $NaOC\ell$, etc.

Since etching time, temperature and concentration of the etchant are all of primary importance, all these variables should be studied individually, keeping the others constant.

CONCLUSIONS

Presently for cosmic ray detection work the thermoset polymer plastic CR39 is the most sensitive material that best satisfies the general preferred properties for such a detection (see Table 2). Although its properties are much superior to other materials, it would be better for the JACEE applications if it could be improved in two respects. Its temperature coefficient of sensitivity for track registration is not ideally small above 0 °C[13]. Its utility in emulsion chamber experiments would be greatly enhanced if it were made somewhat more sensitive. We have suggested approaches of monomer modification, plasticiqer modification, and etchant changes that appear to work in the direction of improved threshold sensitivity. It is likely that simultaneous improvement in track registration temperature stability and post-etching surface clarity may not be achieved with improved sensitivity. A lot of experimental work and calibration with heavy ion beam exposures will be required.

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