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Abstract

It is proposed that the "threshold for etchable track formation" in plastics is associated with the linear density of activated polymeric clusters along the path of an ion, in the "grain-count regime." Existing data for CR-39 reveal a change in etching rate in the neighborhood of z/β having the value of about 15-20. We infer that this defines the transition from the grain count regime to the track width regime, as the activation cross-section exceeds the cross-sectional area of the cluster. This interpretation is consistent with available data for the G value for polymer scission and the dose of gamma-rays required for enhanced etching of the bulk material. CR-39 appears to be a 1-hit detector. A similar track theory interpretation appears to be consistent with observed changes in bulk etching of photoresists irradiated with electrons and heavy ions of different LET.

Introduction

Although 25 years have elapsed since the discovery that the paths of charged particles could be revealed by chemical etching (Young, 1958), there is still no general agreement about the mechanism of track formation nor the appropriate parameter against which to plot etching rate or "threshold" or track diameter. There is no quantitative model which predicts these quantities. The uncertainties in track formation models sometimes lead to unfortunate errors, as illustrated in the studies of the "magnetic monopole candidate" (Price *et al.,* 1978). Here we inquire further into the relationship between the present state of knowledge and the delta-ray theory of track structure (Katz, 1977).

According to the delta-ray theory of track structure, tracks are caused by the interaction of secondary electrons or delta rays, ejected from the medium by the passing ion, with the sensitive elements of the detector. By the word "track" we mean the distribution of activated sites or elements which lead to the observed end-point. The appearance of the track depends on the end-point as well as on the charge and speed of the ion. For etchable plastics, as for emulsion, the character of the track depends on storage time, the composition of the developer, the developing time, and so on, all of which are included in the term end-point. Since the damage from which the track results is produced by secondary and higher generation electrons, as in the case of irradiation with gamma-rays, it is assumed that the detector can be calibrated with gamma-rays, and that the dose-response relationship determined from gamma-ray exposure can be mapped into the region surrounding the ion's path. We interpret the dose-response relation probabilistically, as the probability for activation of a sensitive site at a dose D. The radial distribution of local dose from delta rays, averaged over the sensitive site, then yields the radial distribution of probability for site activation, and hence the appearance of the particle track. When the radially integrated probability is smaller than the cross-sectional area of the site, we say that the activated sites are like "separated beads on a string" and label this the "grain count regime." When the radially integrated probability is greater than the cross-sectional area of the site, the track assumes the appearance of a hairy rope, and we say that we are in the "track width regime." To describe a particle track, the theory requires the number of sites per unit volume, the site size, and the gamma-ray dose-response calibration, as well as the radial distribution of dose about the ion's path.

Some Generalities

Typically we do not know very much about the detectors to which our models are applied. We frequently know virtually nothing about the details of detector architecture, such as the detailed location of sensitive sites or the nature of the interactions which lead to the end-point, and the different pathways between the initial interactions and the final end-point. Our procedure is to try to develop a set of formulas, parameters, and concepts that describe the widest possible number of detectors, and for which the detailed construction and operation of a detector plays a minimal role. Wherever possible, the functional forms of the dose-response relationships are based on simple statistical models, like the cumulative Poisson distribution. The model is based on target theory. The interpretations are probabilistic.

The probabilistic interpretation immediately invalidates the concept of threshold (Katz and Hofmann, 1982) in all detectors to which the model is applied. There can be no thresholds of effect for probabilistic processes based on dose, but only thresholds of perception, in relation to background. When dealing with rapidly varying effects, small changes in dose levels or in LET may induce sufficiently large changes in effect as to create the impression of a threshold. The appearance of particle tracks in emulsion and the observations of alpha particle tracks in cellulose nitrate (Zamini and Charalambous, 1982) confirm this view. The onset of observability for particle tracks in plastics and minerals gives the impression of a threshold. In earlier work (Katz and Kobetich, 1968), we proposed a criterion of minimal dose at a minimal distance (of about 20 Å) as a threshold criterion, and note that the minimal radial distance criterion was subsequently confirmed in mica (Bean *et al.,* 1970), where a minimal track radius of 33 Å was observed.

As an estimate of the radial distribution of dose about the path of an energetic heavy ion passing through water we may take the calculated result to be (Fain *et al.,* 1974)

$$D = 1.3\,(z^*/\beta t)^2 \text{ megarad Å}^2 \tag{1}$$

out to radial distances limited by the kinematics of energy transfers from the ion to the delta rays, and where z^* is the effective charge number, β is the speed relative to light, and t is the radial distance. We note that doses of the order of megarads are achieved at distances of the order of 10 Å. Many plastic materials display an enhanced etching rate of the bulk material, in sodium hydroxide, when irradiated with gamma rays to doses of the order of 10 megarads (O'Sullivan *et al.,* 1982), offering quantitative substantiation to the delta ray damage model of track formation, as revealed by chemical etching.

In the delta ray theory of track structure, it is assumed that the detector consists of sensitive elements, say emulsion grains, the nuclei of biological cells, isolated molecules, or polymeric clusters, that may contain one or several sensitive sites. These sites may require 1-hit or several hits for activation. These differences are accomodated in the theory through its parameters. Depending on the detail required, we need to know the number density of sensitive sites, say the number of grains per unit volume, the number of hits per detector, the dose of gamma rays at which there is an average of one hit per target, the size of the sensitive target, and if there is a complex structure, as in the biological cell, the size of the container in which the targets are located, as in the bean bag model of the nucleus of a biological cell in which the bag is the nuclear membrane and the beans are the targets, perhaps different strands of DNA. The targets may be single or paired, as in the strands of DNA or in some polymers, where damage must be caused in adjoining chains if there is to be subsequent cross linking.

In the theory, one first describes the character of individual particle tracks and then extends the description to include high dose irradiations where there are inter-track effects. Inter-track effects are observed for detectors which can be sub-lethally damaged, as in the case that several hits are required for the expression of an end point but that a smaller number is likely to be generated in a substantial portion of the track of a single particle. This is frequently the case in the grain count regime for biological cells, and for many hit (desensitized) nuclear emulsions. It can also be the case for some photoresists. According to the theory, this is associated with "supralinearity" in the dose-response curve found after exposure to gamma-rays. It is thought to be prerequisite for the observation of an RBE (relative biological effectiveness) greater than 1 with high LET radiations.

The theory of the survival of biological cells after irradiation makes use of these concepts of track structure, including the accumulation of sublethal damage. It has recently been applied to experimental findings with photoresists (Katz, 1982), using the formalism developed for cells. We are thus led to the view that plastics may be considered as an assemblage of polymeric clusters.

We assume that the observed track etch behavior of plastics is consistent with the structure and concepts of track theory.

We assume that the threshold-like character of track etching phenomena is associated with the grain-count regime. The bulk etching rate is then expected to mask the formation of tracks unless the damage sites are sufficiently close together, perhaps until some 20% or more of the clusters through which an ion passes are activated. Typically the probability of activating a site through which an ion passes is given by an expression of the form.

$$P = (1 - e^{-z^{*2}/\kappa\beta^2})^m \tag{2}$$

where κ and m are parameters of the detector, with m being the number of targets or the number of hits in a single target required for inactivation, and

$$\kappa = (E_0 a_0^2)/(2 \times 10^{-7} \text{ erg cm}^{-1}). \tag{3}$$

Here E_0 is the dose at which there is an average of 1 hit per site and a_0 is the radius of the site.

The value of z^*/β for the transition from the grain-count regime to the track width regime depends somewhat on the character of the detector and the criteria we impose. One criterion, appropriate to track etching, is based on the activation cross-section of the detector, found from integration of the radial distribution in the activation probability. We take the transition to occur when the activation cross-section is equal to the cross-sectional area of the sensitive site. Making use of an earlier calculation (Katz, 1977, esp. Figure 12), we identify the transition from the grain count regime to the track width regime in 1-hit detectors as occurring at $z/\kappa\beta^2 = 2/3$. We expect that the etching rate in a 1-hit detector should exhibit a change in slope, when plotted as a function of z^*/β at the transition from the grain-count regime to the track-width regime, and that there should be no saturation in etching rate with an increase in this parameter (z^*/β) except as governed by the limited range in delta-rays, from collision kinematics, at low ion velocities.

We need an additional relationship to establish the consistency of our arguments. We take it that the G value, E_0, and N (the number density of targets) are related through the expression

$$GE_0 = N \tag{4}$$

stating that the number of targets per unit volume N divided by the energy per unit volume at which there is an average of 1 hit per target E_0 is the number of hit targets per unit energy G. We will further assume the target size to be given from

$$N^{-1} = 4/3\, \pi a_0^3. \tag{5}$$

CR-39: Experiment And Theory

It is a substantial task to estimate the radiosensitivity parameters of plastic track detectors from available data. Geometry must play an important part in the etching process, so that the bulk etching rate after irradiation cannot be directly mapped into the region about the ion's path. Nevertheless, crude estimates may be made from available data.

Thus the ratio of the etch rate after irradiation to the etch rate of the unirradiated material is plotted, as a function of gamma-ray dose, by O'Sullivan *et al.* (1982). From this plot we crudely estimate that the characteristic dose at which there is an average of 1-hit per sensitive site is about 7 megarads. The same reference gives a correlation between the parameter J (originally primary ionization, but subsequently modified) for the track etch threshold and the value of G for scission of polymer strands G_s, from which we note that G_s for CR-39 = 10 scissions/100 eV.

According to Rao *et al.* (1981), there is no saturation in the etching rate for particle tracks, as consistent with our view that etching rate is primarily governed by the track width. They plot etching rate vs. z/β, and note a change in the slope of this graph in the neighborhood of z/β having the value 15 to 20. From our previous discussion we infer that κ lies between 330 and 600.

Making use of the data from O'Sullivan *et al.* in our equations (4) and (5) leads to the value: κ = 500.

Results and Conclusions

We have attempted to apply the perspectives of the delta ray theory of track structure to available experimental data on CR-39, with apparent success. Interpreted in this way the several pieces of available data about the track etch process, the etching rate of the bulk material, and the G_s value for chain scission seem to lead to a consistent pattern. The perception of a threshold arises from the competition between bulk etching and etching along the particle's path, when a substantial fraction, say 20% of the polymeric clusters are activated by the ion passing through them. The absence of a saturation in etch rate is related to the track width regime. It would appear that the diameter of the etch pits must also be related to the track width. We cannot state this relationship any more than we can state a relationship between etch rate and cross-section, because of the complexity of the etching process. We note that the cross-section varies as $(z/\beta)^2$, but the excess etching rate varies as $(z/\beta)^3$, in the track width regime, according to Rao *et al.*

We infer that CR-39 is a 1-hit detector. This would help to account for its greater sensitivity relative to other track etch detectors. If, as suggested by Rao *et al.*, particles with z/β of about 8 can be detected, we would expect to be able to observe the tracks of electrons having an energy of about 4 keV. But such electrons have a range of less than 1/2 micron, suggesting that the bulk material will be etched away before the track is large enough to observe in an optical microscope. Such tracks should be sought in an electron microscope, to test the hypothesis that CR-39 is a 1-hit detector.

Alternatively, we should seek to observe the tracks of muons and pions.

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