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A.M. Bhagwat

Solid State Nuclear Track Detection: Theory and Applications

**Indian Society for Radiation Physics
Kalpakkam Chapter
1993**

About ISRP and this Series

Ionising radiation is a powerful tool finding increasing applications in almost all walks of life, be it agriculture, medicine, industry or basic research. By their very nature of its diverse applications, the study of ionising radiations and their interaction with matter has diffused into various other scientific disciplines. It is with primary objective of providing a common forum for the scientists and engineers working on different basic as well as applied aspects of ionising radiation that the Indian Society for Radiation Physics (ISRP) was formed in 1976. Its membership consists of professionals from national laboratories, universities and institutions of higher education, industry etc. In line with its basic objective, ISRP has been organising periodic national and regional seminars, topical meetings etc.

It is recognised that for an optimum utilization of any technology, a comprehensive appreciation of its problems and potentials must prevail not only amongst the scientists and engineers associated with the technology but amongst the general public also. In the case of ionising radiations while its hazard aspects seem to have been over played for historical or other reasons, its full potential in the service of mankind does not seem to have drawn the deserved attention of the general public. It is to fill up this gap and to develop an overall perspective, ISRP (Kalpakkam Chapter) has launched this series of semi-popular brochures and technical reviews on various facets of ionising radiation.

We feel that for any programme to be relevant and successful, a strong user-feed back is essential. We earnestly solicit suggestions with regard to the content and level of these brochures, topics to be included etc. The suggestions may please be sent to:

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FOREWORD

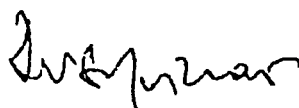
Ionising Radiations and radioactive materials are increasingly assuming major role as useful tools in the service of mankind. However, mis-apprehensions about the harmful effects of ionising radiations at any level have also grown to large proportions in the recent past. Unless conscious efforts are made to build up a proper awareness and control this syndrome, a point may soon be reached where the benefits may get totally marginalised by undue fears.

There is no denial of the fact that uncontrolled radiation exposure is harmful to life. But adequate scientific information on radiation protection is available today that permits us to derive benefits from radiation without incurring any harm. What is needed most is presenting the facts in proper perspective through dissemination of scientific information on ionising radiations to all sections in the society. It is in this context that the inadequate scientific literature at the semi-technical and common man's language has been strongly felt over the past decades. To fill this lacuna the Kalpakkam chapter of Indian Society for Radiation Physics(ISRP) has taken upon itself the task of bringing out appropriate literature in the fields related to ionising radiations. The efforts made so far by the Kalpakkam Chapter have already borne fruit and the monographs/booklets brought out over the last five years have been eminently useful.

The continuing role of radiation detection and measurement techniques in quantifying the ionising radiations can never be over-emphasised. New developments in this field must therefore be welcomed and given due recognition. This monograph on "Solid State Nuclear Track Detection (SSNTD): Theory & Applications" by Dr. A.M. Bhagwat which introduces a new technique in radiation detection & measurement is an effort in this direction. An important feature of this technique is its inexpensive nature and as such amenable for use in the universities and colleges. The text includes a list of simple experiments that can be conveniently

introduced at the undergraduate/post graduate level. It is worth noting that like other techniques of radiation detection & measurement, this too has a multidisciplinary outlook and potential.

I consider it my pleasant duty to put on record my sincere appreciation of the efforts put in by the author and the Kalpakkam Chapter of ISRP in bringing out this monograph. I wish this programme of ISRP all success.



(D.V. GOPINATH)

Director

Health, Safety and Environment Group
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SOLID STATE NUCLEAR TRACK DETECTION: THEORY AND APPLICATIONS

1. INTRODUCTION

The field of Solid State Nuclear Track Detection(SSNTD) had its origin in the year 1958 when first observations were reported by D.A. Young at AERE Harwell (Young, 1958). He noticed that when lithium fluoride crystals, held 1 mm away from uranium oxide film, were irradiated with thermal neutrons the crystal surface revealed a number of shallow etch pits after treatment with a chemical reagent ($\text{HF} + \text{CH}_3\text{COOH}$), saturated with FeF_3 . Further the number of these pits (as seen under the optical and electron microscope) had a one to one correspondence with the theoretically estimated number of fission fragments recoiling into the crystal from the uranium foil. A particle detector was thus born but its potential was not realised until 3 years later. In his short paper in Nature (Young, 1958) Young had recognised the existence of tracks, demonstrated that they could be etched and made optically visible and explained their formation as resulting from the trails of damage left behind by the passage of fission fragments.

The direct observation of these damaged regions, produced by fission fragments, in thin sheets of mica, under transmission electron microscope was reported in 1959 by E.C.H. Silk and R. S. Barnes (Silk & Barnes, 1959) also of AERE Harwell. They were unaware of the work published by D.A. Young.

The credit for the development of a new particle detector, from above observations, goes to R.L. Fleischer, P. B. Price and R. M. Walker who began in 1961 where Silk and Barnes left off. Since then, the field has grown to such an extent that now there is hardly a field of science and technology where it does not have an actual or potential application. To learn something about such a subject, which has a multidisciplinary outlook and potential, will not therefore be out of place. This booklet provides a bird's eyeview of the subject and is intended to serve both the beginners and specialists alike. The readers interested in digging deeper, may find more extensive reviews of the subject in several books: Becker, 1973; Fleischer, Price & Walker, 1975; Monnin 1978; Durrani & Bull, 1985; Griffith & Tommasino, 1990.

In the following sections, we describe the two main steps involved in SSNTD, viz. the formation of latent tracks and their subsequent development by chemical or other means. These two steps are reminiscent of similar stages in photographic emulsions, but the similarities end there. The physical mechanisms involved in the two phenomena are quite different, as will become clear from the descriptions to follow.

No book on SSNTD can be complete without reference to its variegated applications. We shall touch upon these in the concluding phases of this book.

2. A CHRONOLOGICAL ORDER

The observation of charged particle tracks did not begin with the discovery of SSNTD phenomenon. They were first made visible in the cloud chamber (due to charge induced nucleation) and then followed by bubble and spark chambers. These methods however suffered from one major drawback viz. the tracks could not be retained permanently. Next in line came the nuclear emulsions which could retain charged particle tracks but they could not be handled freely and with ease. The appearance of the SSNTDs on the scene removed this last difficulty also.

The discovery of each of the above detector types may be either accidental or by design. But the chronology of events imparts a logic to scientific efforts which may otherwise get classified as accidents. A chronological order is thus easily seen in the above developments.

3. LATENT TRACK FORMATION

Charged particles passing through dielectric material (crystals, inorganic glasses & plastics) produce submicroscopic trails of continuous damage along their path. These trails consist of radiation damaged material and are called latent tracks as they are too small to be seen even with the help of a microscope. Under favourable circumstances, they can be viewed under Transmission Electron Microscope (TEM).

Over the years, the characteristics of charged particle tracks have been studied in several insulating materials. Some of the salient features are as follows:

i) Tracks are regions of damage composed mainly of displaced atoms rather than electronic defects. They are stable, chemically reactive centers of strain.

ii) The region of damage is narrow, i.e. less than 10 nm in diameter and continuous atomically or nearly so. The length of damage trail is equal to the range of the charged particle in the medium under consideration.

iii) Tracks are not formed in metals and good semi-conductors. Table 1 indicates the categories of track-storing and non track-storing materials. We see a clear correlation between the electrical resistivity of a material and its ability to store tracks. In general, only materials having resistivity values greater than 2000 ohm.cm store tracks.

iv) No track formation occurs, unless the rate of energy dissipation by the charged particle exceeds a critical value. This value is different for different detectors. As a result, there is a gradation of sensitivity among the solids that record tracks.

Table 1

Relation of track formation to electrical resistivity

Materials	Resistivity range (ohm.cm)
1.Track forming	
Insulators:Silicate minerals	$10^6 - 10^{20}$
Alkali halides	
Insulating glasses	
Polymers	3000-25,000
Poor insulators: MoS ₂	
Semiconductors : V ₂ O ₅ glass	
2.Non track forming	
Semiconductors: Germanium	10 - 2000
Silicon	$10^{-6} - 10^{-4}$
Metals: Al, Cu, Au, Pt, W, Zn.	

4. MECHANISMS OF LATENT TRACK FORMATION

Many models have been proposed to explain the mechanism of track formation. We shall discuss the ones which are consistent

with the characteristics of the tracks described a little earlier. Let us take the case of inorganic solids first because they account for about 80% of the track detectors studied and their case is also simple.

4.1. Ion Explosion Spike Model

This model was put forward by Fleischer and Price in 1965 and appears to be a working theory concordant with experimental results. It takes into account the extreme insensitivity of the inorganics to electron bombardment. According to this model, the burst of ionisation along the path of a charged particle creates an electrostatically unstable array of adjacent positive ions which eject one another from their normal sites into interstitial positions as a result of Coulombic repulsion. Fig. 1 illustrates this multistep process.

Following primary ionisation (step 1) which takes place in less than a femto second, an array of interstitial ions and vacant sites is created (step 2). After this, elastic relaxation takes place which diminishes the acute local stresses by spreading the strain more widely. It is the creation of these long range strains (in step 3) which makes possible direct observation of unetched tracks in crystals by transmission electron microscopy.

This model holds ionisation to be almost exclusively responsible for the formation of tracks in insulators. Two observations strongly favour this point of view. First, the tracks are observed at practically full length whereas atomic collisions principally occur at the end of the path.

Secondly, no tracks are formed in conductors or semi-conductors where only atomic collisions are able to produce stable defects. In such solids, ionisation is indeed a transient phenomenon since the number of free electrons is large

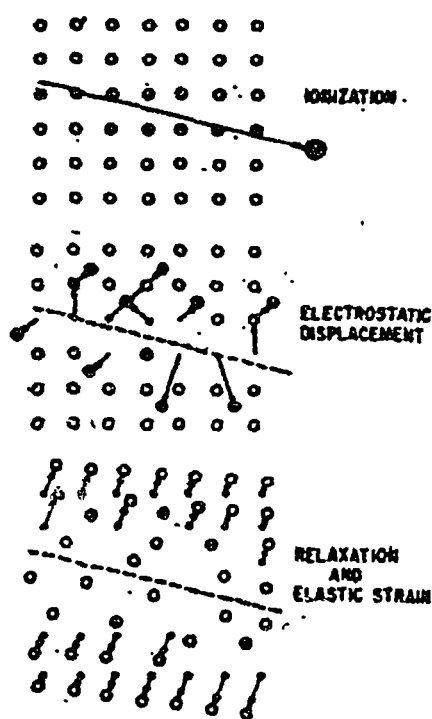


Fig.1 Ion explosion spike mechanism.

enough to compensate very quickly any electronic defect.

Evidence is strong that the secondary effects of delta rays and excitation processes are unimportant in inorganic solids.

4.2. Radiochemical Damage Mechanism

The case of organic solids (polymers) is in sharp contrast with that of the inorganic solids, in that the effect of delta rays cannot be neglected. This is so because polymers are severely damaged by electron bombardment at doses of about 100 kilo Gray which are several orders of magnitude lower than doses required to produce observable damage in inorganic material, i.e. 10 giga Gray in silica glass to loosen silicon oxygen bonds. Further, in polymers, the deexcitation following an excitation process can lead to breaks in long chain molecule and to free radical formation as only few eV of energy are required for producing such changes. Track formation in polymers is therefore based on radiochemical damage mechanism described below:

According to this model, etchable tracks are formed by radiolytic scission of long polymer chains into shorter fragments and the production of reactive low molecular weight radiolytic products which are more easily dissolved by etchants than the surrounding undamaged plastic.

Thus, both primary and secondary ionisation and excitation processes must be considered while explaining track formation in organics. Fig. 2 illustrates this process.

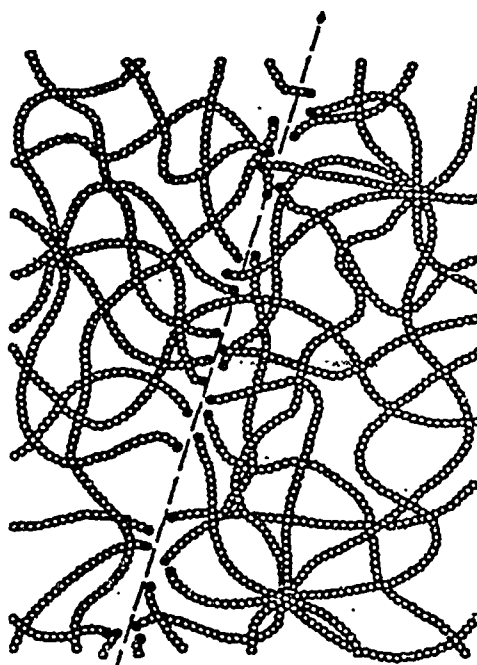


Fig.2 Radiological damage mechanism.

5. REQUIREMENT FOR LATENT TRACK FORMATION

The search for possible SSNTD materials can be considerably simplified if the physical and chemical properties of the medium favouring the formation of tracks are identified, preferably in a

quantitative fashion. Mechanistic models described above provide a logical insight into this aspect.

5.1. Inorganic Solids

i) In order for the ion explosion to take place the pressure due to Coulomb repulsive forces inside the ionised channel must be greater than the coherent attractive forces due to lattice bonding, i.e. local electrostatic stress must be greater than the local mechanical strength or bonding strength. This condition tells that tracks should be formed easily in materials of low mechanical strength, low dielectric constant and close interatomic spacing. It also justifies why plastics are more sensitive than glasses which are more sensitive than most crystals.

ii) The positive ions produced in the channel should not get neutralised within 100 femto seconds (lattice vibration time) because approximately so much time is required for the ions to be displaced from their lattice sites. This is realised only in those solids which have a conduction electron density of less than $10^{20}/\text{cm}^3$. This implies that tracks will not form in metals, as was pointed out already.

iii) To maintain permanently the domains destroyed by the charged particle, the hole mobility in the detector substance must also be low, i.e., less than 0.1 m/s for a field strength of 100 volt/m . This requirement excludes numerous semiconductors from the list of possible detector substances.

iv) Developable areas destroyed by the charged particle must be continuous atomically (or nearly so), i.e. there must be at least one ionisation per atomic plane crossed by the particle. This in turn depends upon particle parameters (its type and energy) and the ionisation energy of the detector substance (this criterion applies to chemical etching technique). It follows therefore that each detector system should have a different threshold of detection. We shall discuss detection thresholds and etchability aspects in more detail in a later section of this book.

5.2. Organic Solids

While it is well known that organic solids including plastics have a low detection threshold, quantification of criteria has not been possible as in the case of inorganic solids. The picture remains essentially qualitative.

Little is known at present about the relationship between physi-

cal and chemical properties of a material and its capability to register tracks. It is therefore difficult at this stage, to predict the sensitivity of a new material. However, some generalisations can be made:

i) Systems that are highly unstable chemically, such as cellulose nitrate (CN), tend to be more sensitive than radiation resistant polymers.

ii) Thermoset materials like CR-39 that do not cross-link upon irradiation and are susceptible to interfacial degradation by a convenient etchant should constitute an ideal polymer detector. Their amorphous nature and radiation sensitivity will further enhance the track detection property. Introduction of weaker linkages in the polymer structure, that are easily broken by charged particles, will further enhance the sensitivity as in case of SR-86. (Fujii et al, 1990).

6. TRACK VISUALISATION TECHNIQUES

Since the latent track diameters are less than 10 nm, their direct observation calls for a Transmission Electron Microscope (TEM). Although TEM has proved very useful in the examination of shape of tracks and diameter of the damaged region, it has many serious limitations for regular use with SSNT detector. For instance, only very thin samples (a few tens of nanometers in thickness, which are not easily obtained, can be used. Further, the observation surface is very small, hence the necessity for very high track densities (of about 100 million per square centimeter or more). Moreover, it is difficult to see the tracks at full length (of the order of 10 μm). In some materials tracks are annihilated upon the impact of electrons. For practical purposes therefore, some type of track development is necessary which results in their enlargement to sizes that can be seen under an optical microscope. The following method serves this purpose.

6.1. Selective Chemical Etching

The damage trails represent regions of enhanced chemical activity compared to bulk material. This is because they consist of disordered structure which in turn are associated with large free energy. In this method the detector material containing the damage trails is immersed in a suitable chemical solution. The damage trails which intersect the surface get preferentially dissolved in the etching solution, compared to the bulk material. When their dimensions become comparable to (or greater than) wavelengths of visible

light, they act as strong scattering centres appearing black in normal bright field illumination and white in a darkfield under the microscope.

This process of enlarging track dimensions by chemical action is called chemical etching. The etching reaction is a heterogeneous process involving a solid liquid interface. The driving force for the reaction is the reduction in the free energy of the system.

Evidently, for the formation of the track by chemical etching, the rate of etching along the track " V_T " must be greater than the rate of etching of the bulk material " V_G ", i.e. V_T/V_G should be greater than one. This ratio is an important parameter and denotes the degree of preferential etching along the particle trajectory.

Here it should be remembered that it is the observed track length which increases as the etching proceeds. The etchable track length, however, remains constant. In fact, this length tends to decrease as a consequence of surface erosion of the insulator. Secondly, after an incubation period, necessary for the penetration of the etchant and dissolution of the intensely damaged core region, the transverse dimensions of the track vary linearly with time.

A simple chemical etching apparatus is shown in fig. 3.

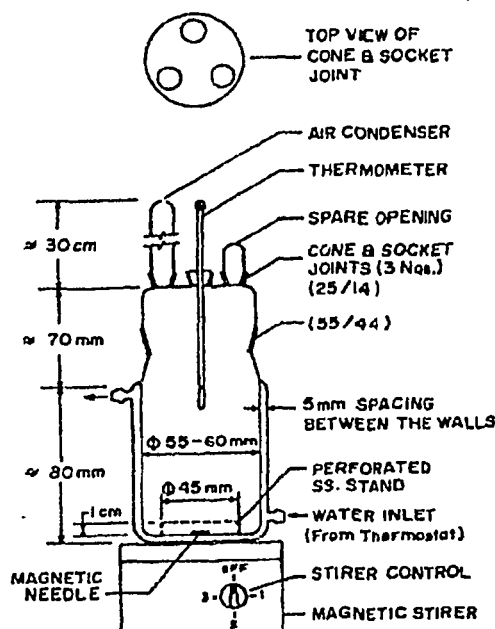


Fig.3 Simple chemical etching apparatus.

6.2. Electrochemical Etching (ECE)

Among the various methods which have been proposed to enhance the etching speed and/or the size of the etched region, one which has become promising is electrochemical etching. In this technique, chemical etching at tracks is combined with an electrical breakdown process called treeing, observed in the bulk of in-

ulators.

At the beginning of etching, the reagent rapidly diffuses in the pre-existing latent tracks, turning them into conductive paths which penetrate the insulating foil. If an alternating voltage is now applied through the dielectric (during etching), currents can be produced in these conductive paths. The associated conductive energy losses in track regions, which are large at high frequencies and high voltages, lead to local heating of the etchant (the dielectric specimen being unable to dissipate heat from the very localised regions). This in turn increases the etching rate V_T . Further, at these high voltages, large field concentrations arise at the tip of these paths which are conducive to development of treeing process and provide a way to enlarge the damaged track regions at will, by allowing V_T to increase. The rise of temperature and the stress concentration at the end of the conductive channels produce failure of the dielectric, i.e. its thermal degradation. This leads to continuous growth of current and complete degradation along the path reducing the tracks to nothing but carbonised paths. This is why ECE tracks are seen as black spots under the microscope.

The conduction currents due to absorbed ions and other forms of dielectric losses generate heat continuously in the electrically stressed insulators. This may raise the temperature of the dielectric foil and thus increase the chemical action of the reagent as a whole, i.e. increase both V_G and V_T .

This technique has been certainly found to be superior to conventional chemical etching because it permits such a large scale amplification (20,000 for tracks of 200 μm diameter, assuming an original latent track diameter of 10 nm) of the tracks that they can be either seen by the naked eye or can be projected on a screen with a slide projector (or a microfiche reader). A ten film ECE cell is shown in fig. 4.

The ECE technique has mainly been found to work with polymers with following general properties (Bhagwat, 1983):

- i) Good electrical properties, i.e. high values of tensile strength, dielectric strength and volume-resistivity,
- ii) Low water absorptivity,
- iii) Low value of dissipation factor or dielectric loss factor, and
- iv) Polar nature of the material.

Polymers like Lexan, Makrofol and CR-39 have been found to give good result in ECE.

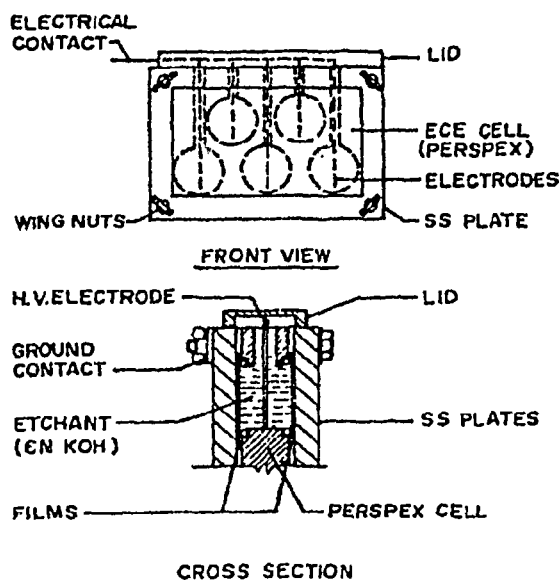


Fig.4 Ten film ECE cell.

it here, as it has not found much practical use. Tracks developed by various techniques are shown in fig. 5.

ECE has been found to have lower efficiency than CE. However, a combination of CE and ECE leads to improvement in efficiency. This combination is now being employed in many applications. Recent efforts with high temperature ECE indicate that prior chemical etching may not be required.

There is another technique of track revelation known as Graft and Dye technique. We will not discuss

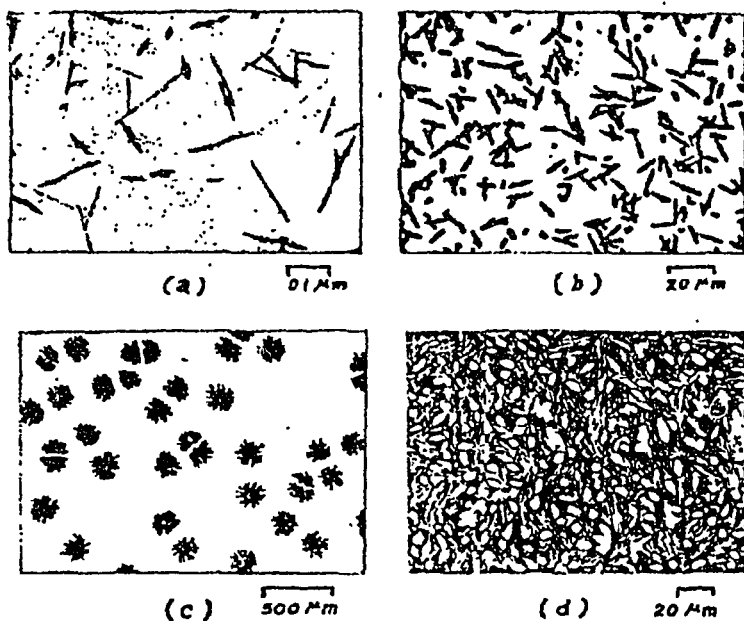


Fig.5 Tracks developed by various etching techniques.

7. CRITERIA FOR TRACK REGISTRATION BY CHEMICAL ETCHING

Not all the latent tracks which intersect a given detector surface may lead to etchable tracks. Obviously, etchability of tracks depends on particle parameters like charge Z , mass m and velocity v . Several criteria have been proposed to find an appropriate form for this functional dependence on Z, m and v , which will best predict the realisation of the condition $V_T/V_G > 1$. A simple and valid track formation criterion must have some parameter X which will have unique threshold value X_c for a given detector. This value X_c of the parameter will separate clearly the region of track etchability from that corresponding to non- etchability.

In recent years this view of track formation has been modified somewhat in that less emphasis is placed on threshold values of X . Experimental data is now available which shows that track etching rate V_T is a smoothly varying function of incident particle parameters. The parameter X is therefore so chosen that $V_T(X)$ has always the same value (for a given detector and etching conditions) at a given value of X regardless of combination of ionic charge and particle velocity that may lead to X value in question. In other words, the curve of V_T versus X is independent of the particle type. Therefore, once $V_T(X)$ is determined, etching rate of any ion at any energy may be calculated. Correlations of this nature, however, put restrictions on track formation mechanisms that can be put forward. Some of the criteria that have been proposed so far are described below in brief.

7.1. Total Energy Loss Rate (dE/dx) Criterion

This was the first criterion proposed and probably offered the most natural explanation (Fleischer, Price, Walker & Hubbard - 1967a). It prescribed that for etchable track formation; the total rate of energy loss per unit path length by the bombarding particle must exceed a critical value which is characteristic of the particular detecting material. The particles losing energy more rapidly than this value would produce continuous tracks with unit efficiency while those lying below this threshold will produce no tracks. This criterion seemed reasonable in the initial years when limited track registration data was available. However, it failed when applied to relativistic particles. The reason being that relativistic bombarding particles produce energetic delta rays which tend to be multiply scattered away from the latent track region depositing their energy

away from the path of the primary particle i.e. energy loss rate also includes energy not deposited in the track region. The criterion was therefore rejected by its propounders (Fleischer, Price, Walker & Hubbard 1967b) in favour of another criterion described below.

7.2. Primary Ionisation (J) Criterion

It was argued that a quantity somewhat different from energy loss rate should determine the presence or absence of etchable track. According to this new criterion, which is very much in tune with the ion explosion spike model, the track formation should depend on number of ions formed per unit distance close to the particle trajectory i.e. a dielectric solid will record a track if the rate of primary ionisation exceeds a critical rate characteristic of the material.

The parameter J, representing the primary ionisation rate or density of radiation damage, is a function of atomic number "Z" and velocity " β (v/c) of the particle. A functional form based on the work of Bethe is usually employed.

$$J = \frac{a \cdot Z_e^2}{\beta^2} \left[\ln \left(\frac{\beta^2}{1 - \beta^2} \right) + K - \beta - \delta(\beta) \right]$$

where Z_e = effective charge of the particle

a and K = constants for a given stopping medium.

$\delta(\beta)$ = Relativistic correction term to account for the polarisation of the stopping medium by the electromagnetic field of the moving particle (for $\beta < 0.8$, $\delta = 0$).

This equation accounts for all the etching rate measurements made thus far. For a given solid-etchant combination the constant "K" should be so chosen that the etching rate depends on "J" only within experimental errors. It has not yet been possible to calculate K from first principles or to make an absolute determination of J for most stopping media. Further, the ionisation potentials implied by values of K (used in obtaining the best fit to experimental data) are physically unrealistic. Therefore, according to authors, the above equation should be regarded at present more as a useful phenomenological equation than as one of deep theoretical significance.

Notwithstanding these limitations, the central idea behind this criterion, i.e. primary ionisation processes should predominate over

secondary ones as the basis for track formation, appears reasonable considering that in crystals the radial extent of unetched tracks is only a few nanometers. This criterion has proved highly successful in practice except that the concept of threshold J is now-a-days regarded as less decisive. For both plastics and minerals, the track etching rate V_T is a continuous function of J . Typically V_T is proportional to J^α where $\alpha \simeq 2$ has been found to hold for some plastics. (V_T versus J curves generally rise very steeply).

7.3. Restricted Energy Loss (REL) Criterion

This criterion (Benton, 1967) takes into account the secondary ionisation and excitation (in addition to primary processes) produced by low energy δ -rays in the region of track core i.e. it makes allowance for the energy not deposited in the track core by high energy δ -rays. The main underlying assumption here is that the total energy deposited in the core region by the incident charged particle determines the chemical reactivity (i.e. the etchability of the latent track region. Thus a particle will produce an etchable track over the extent of its range where REL is greater than a threshold value $(REL)_{crit}$ for the material.

Calculations are made by selecting a cut off value W_0 of energy of δ -rays such that only those δ -rays whose energy is less than W_0 contribute to track formation. For the purpose of fitting REL to experimental data, W_0 has been assigned values of 200, 350 or 1000 eV. The quantity $(dE/dx)_{W < W_0}$ varies with ion energy in a manner rather similar to primary ionisation and gives a good fit to experimental data. Further REL criterion gives values of $(dE/dx)_{W < W_0}$ or $(REL)_{crit}$ in absolute units.

The choice of W_0 as a fitting parameter introduces certain arbitrariness into REL calculations. Electrons with $W < W_0$ also lose part of their energy in track region which is not included into REL. Similarly electrons with $W < W_0$ lose part of their energy outside the core region. This should be excluded. The physical basis of W_0 is therefore not fully clear.

7.4. The Delta-ray Criterion of Katz and Kobetich or the Secondary Electron Energy Loss Criterion

This approach (Katz and Kobetich, 1968) conflicts directly with the criteria 7.1 and 7.2 described above. This proposition suggests that energy deposited by δ -rays, rather than primary ionisation events themselves, is crucial in the formation of etchable tracks (in

case of α -particles, secondary ionisation accounts for 60-80% of total ionisation). It is proposed that etchable track damage results when a critical dose (D_{crit}) of ionisation energy is deposited by secondary electrons at a critical distance from the ion path. The critical distance is taken to be approximately 2 nm as is appropriate for the passage of etchant along the track and diffusion of reaction products back to the surface. It is postulated that at the critical radius, the dose required for etchable damage production approximates doses producing observable bulk damage due to gamma irradiation. By plotting track etchability data on curves of "dose at 2 nm" Vs ion energy, self-consistent dose thresholds can be drawn which separate the etchable and non—etchable regions. These thresholds are about 350 kGy for muscovite mica and 25 kGy for CN.

Although this criterion can also be fitted to most of the experimental data, it is difficult to evaluate this approach in detail because of the following :

(i) It appears unreasonable that the contribution of primary ionisation can be completely neglected.

ii) There is uncertainty about the exact form of range-energy relations for electrons in the eV region.

iii) Other assumptions in the calculations such as the form of angular distribution of ejected electrons are not realistic.

Further, it has been noted that according to this criterion minimum detectable charge for relativistic ions in Lexan plastic is 70 while experimentally this charge has been found to be as low as 57 ± 2 .

7.5. Delta-rays Criterion of Monnin

The approach (Monnin, 1968) is similar to that given in criterion 7.4 above. However, no minimum critical radius is assumed initially. Dose calculations are made within cylinders of increasing radii. It is then postulated that etchability is created when the absorbed dose exceeds a specific value within a cylinder of certain radius. Calculations show that dose falls off quickly as a function of distance from the trajectory. These calculations need refinement to include electron multiple scattering and angle of emission of electrons.

This model also faces the criticism as given in criterion 7.4 above.

7.6. Radius Restricted Energy Loss (RREL) L_r Criterion

This criterion differs from REL criterion of Benton described above in the sense that it includes energy deposited by all the events occurring within a radius 'r' of the particle trajectory (Paretzke, 1977). Thus L_r criterion gives importance to the track region and attempts to consider only that part of energy of δ -rays which gets actually deposited in the core region irrespective of the energy of the δ -rays. L_r is given by

$$L_r = L_{\infty} \frac{a \cdot Z_{eff}^2}{\beta^2} \left[\ln \frac{R}{r} - \left(1 - \frac{r}{R} \right) \right]$$

where L_{∞} = total rate of energy loss dE/dx
 R = maximum track width
 a = constant for a given medium
 r = radial distance from the particle trajectory,
an adjustable parameter in the model.

This model is an improvement over Benton's criterion.

7.7. Lineal Event Density (LED) Criterion

Lineal event density (Paretzke, 1977) is the total number of events occurring within a distance 'r' of the track core which consist of primary and secondary ionisations as well as excitations. This quantity, like primary ionisation of Fleischer et al, is rather difficult to calculate but should lead to more accurate representation of the track formation process.

7.8. A general comment on models

Tracks are formed by ions at energies for which electronic interactions are the dominant mode of energy loss. This is an important constraint on the acceptable models of track formation. (Nuclear interactions become important at the very end of the ion's path where etchable damage is usually not produced).

8. CRITICAL ANGLE OF ETCHING

Assuming that the charged particle parameters and the detector characteristics are such that an etchable track can result, its actual observation is influenced by a geometrical factor described as "critical angle of etching." It is characteristic of each detector-etchant combination and also depends on etching temperature and is explained in fig. 6. Let us consider a track lying at an angle θ

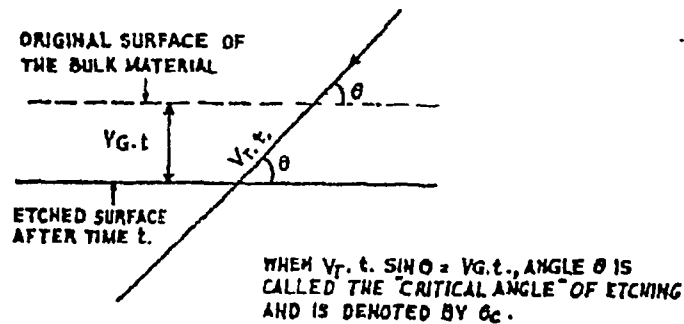


Fig.6 Critical angle of etching.

with respect to the detector surface which has been etched for time 't'. Then the track length etched = $V_T.t$ and bulk layer removed is $V_G.t$. The track will then be observed if vertical component of $V_T.t$, i.e. $V_T.t \sin \theta$ is greater than $V_G.t$. Taking the condition $V_T.t \sin \theta = V_G.t$ and denoting θ by θ_c under these conditions, it is seen that $\sin \theta_c = V_G/V_T$ or $\theta_c = \sin^{-1} V_G/V_T$. The angle θ_c is then called critical angle of etching. Because of this directional characteristic of the detectors, their detection efficiency "is less than unity and can be shown to be equal to $(1 - \sin \theta_c)$ or $1 - V_G/V_T$. Because of this limitation, these detectors are equivalent to other conventional detectors (as regards their detection efficiency) only if the angle of incidence of charged particles in the two cases is equal to or greater than critical angle of etching θ_c .

The value of θ_c in ECE process is still larger, which results in much lower track detection efficiencies.

9. DETECTION THRESHOLDS OF COMMONLY USED TRACK DETECTORS

The detection thresholds of different track detectors vary considerably and depend upon the atomic number (Z) and energy (E) - or more precisely, the velocity (v) - of the charged particle. A parameter Z/β (where $\beta = v/c$) is normally used to characterise the minimum detection limit of a detector; the lower its value, the more sensitive is the detector. Its values are 6, 30, 40 and 60 for CR-39, Cellulose Nitrate (CN), Cellulose Acetate (CA) and Lexan respectively. The order of decreasing sensitivity is CR-39, CN, CA, Lexan, Chronar, Melinex, Mica and meteoritic minerals; these thresholds are shown in fig. 7. The detection thresholds of these detectors can be specified in terms of their energy loss rates, as shown in Table 2.

Table 2

Detection thresholds of some detectors

Detector	Detection Threshold	Remarks
Inorganics	— 15 MeV/mg. cm ⁻²	—
Organics (Lexan, Makrofol etc.)	— 4 " "	—
C.N.	— 1 " "	—
CR-39	< 0.05 " "	(\approx 100 times more sensitive than polycarbonates like Lexan)
SR-86 (It is CR-39 containing sulphonate linkages to a certain extent)		Three times more sensitive than CR-39 for alphas and high energy heavy ions.

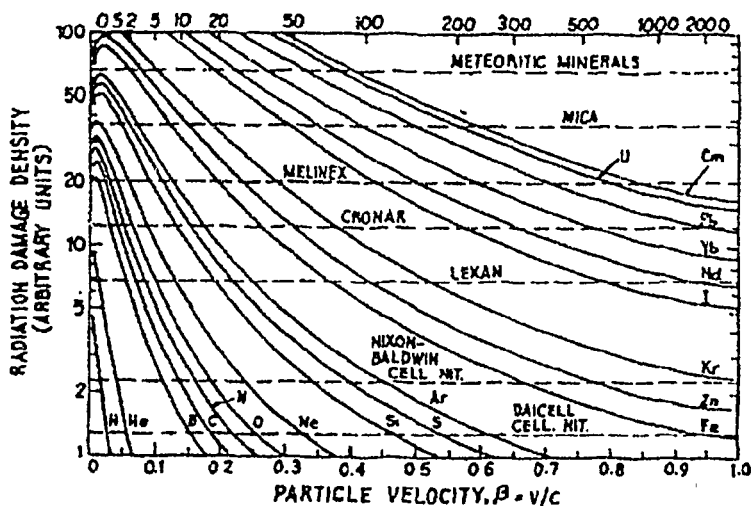


Fig.7 Detection thresholds of common SSNTDs.

10. SOME USEFUL FEATURES OF TRACK DETECTORS

- i) They are inexpensive, convenient to use and quite robust.
- ii) They can be obtained in any size, from very small to very large (small detectors can be used to measure particle fluxes in odd locations while large detectors are used to record very rare events in cosmic ray studies).
- iii) The registered tracks are a permanent record of the phenomenon under investigation. In particular, they remain unaffected by changes in atmospheric conditions such as temperature, pressure, humidity etc.
- iv) They are insensitive to light. Their development or etching is simple and rapid and does not require dark room facilities.
- v) They are insensitive to β , γ and X-radiation.
- vi) They can be used as threshold detectors, e.g. glasses and certain plastics record fission fragment tracks but do not record alpha particle tracks.
- vii) The charge and energy discrimination of these detectors has been found to be better than that of nuclear emulsions.
- viii) Rapid and automatic techniques can be employed with these detectors to count the number of events occurring.

11. APPLICATIONS OF TRACK DETECTORS

The intrinsic features of SSNTDs like passivity, availability in any size, their remarkable stability have contributed to their application in a wide range of fields, often opening up new vistas which were practically unthought of a decade or two ago. The number of applications is too many to be listed in a book of this size. We cite just a few to bring out the versatility of the technique. We limit the descriptions to a bare outline of the methods.

11.1. Applications to Nuclear Physics

Much of the use of SSNTD in nuclear physics research comes from the ability of the method to identify the type of charged particle that produced the track. In the case of an etched track, two parameters are essentially measured: track etch rate V_T and the residual range R at which V_T holds good. R is a function of charge, mass and velocity of the particle while V_T is related to dE/dx of the particle, i.e. to particle charge and velocity. After making measurements on V_T and R at several points along the track they are compared with standard curves for known particles to identify it.

Some other typical nuclear processes studied are indicated below.

11.1.1. Studies on fission phenomenon

After exhaustively investigating the phenomenon of binary fission, the track technique is now being used to study ternary and quaternary fission of heavy (e.g. Uranium) elements induced by heavy ions such as ^{84}Kr , ^{136}Xe or ^{238}U of high energy. These reactions are being interpreted in terms of sequential fission of excited reaction products.

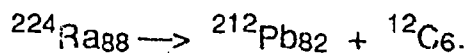
11.1.2. Search for super heavy elements

Track detectors are being employed in the search for any naturally occurring super heavy elements as also in experiments designed to create them artificially within the island of stability around $Z=114$, e.g. by bombarding ^{248}Cm targets with ^{48}Ca projectiles. The superheavies are expected to undergo alpha and spontaneous fission decay associated with larger energy releases, i.e. their alphas and fission fragments should be identifiable on the basis of their longer track lengths.

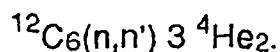
11.1.3. Other phenomena

Other processes being studied include

- i) Study of extremely rare modes of radioactive decay, e.g.



- ii) Study of specific nuclear reactions like



11.2. Elemental Content and its Distribution

This application is a natural extension of the use of SSNTD for particle identification. The method is based on emission of a charged particle from any nuclear reaction such as (n,f), (n, α), (n,p) etc. A number of elements have been studied in this way, e.g. Li, B, Pb, Bi, Po, Th, U, Pu etc. The registered tracks give information on both total elemental content and its spatial distribution; the resolution in mapping depends on the ranges of emitted charged particles, smaller the ranges better the resolution.

If the nuclear reaction produces an alpha emitting radioisotope

of sufficient half life, track detector can be exposed after irradiation is over. It avoids irradiation of the detector by primary particles and also its exposure to high temperature environment during irradiation.

A recent application of uranium fission autoradiography is investigation of "soft errors" in silicon memory devices. The cause of these errors is alpha radioactivity from minute amounts of U and Th contamination in these devices. The passage of alpha particle produces sufficient electronic charge to temporarily alter the contents of a bit location in a memory area. Technique is very sensitive as amounts of U as low as a picogram corresponding to an alpha particle flux of $10^{-3} \text{ cm}^{-2} \text{ h}^{-1}$ could be detected.

Charged particle radiography, using these detectors, is another proliferating technique offering innumerable possibilities.

11.3. Dosimetric Applications

Applications to dosimetry bank on the ability of SSNTD to integrate the signal over a significant length of time. The charged particles tracked are specific, usually alpha particles or recoil protons. Some common instances are:

11.3.1. Inhalation of alpha active aerosols

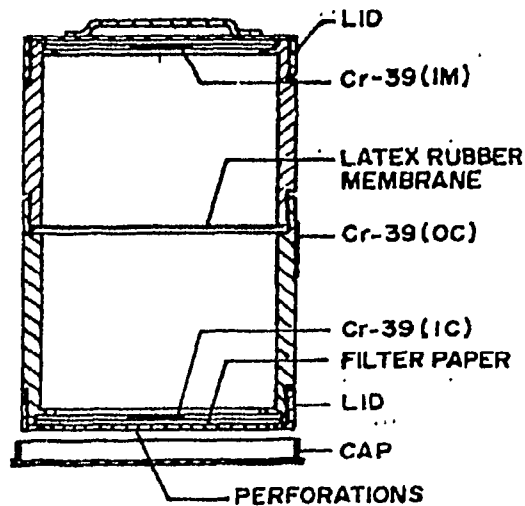
Keen interest has been taken in recent years in the radiological consequences of inhalation of alpha active particles. This is reflected in the intensive measurements presently being carried out in dwellings, mines and caves to assess the concentration of radon and its daughter products. Their deposition and microdistribution in lungs is now being thoroughly investigated for estimating radiation dose to various parts of lung. Use of electrets in improving the sensitivities of SSNTD technique in this area is being presently explored. A radon dosimeter cup presently in use in BARC is described in the following paragraphs.

11.3.2. Radon dosimeter

The dosimeter consists of a cylindrical aluminium cup open at one end. This cup is partitioned into two equal compartments by a thin latex rubber membrane. Three SSNTD films are required for a full measurement. Two films are mounted inside the cup, one in each compartment, and the third film is mounted on the outside surface of the cup.

Radon and Thoron gas enter the first compartment through a filter paper covering the perforated open end of the cup. The gas

subsequently enters the second compartment through the membrane partition. No daughter products pass through either the filter paper or the membrane. The fractions of radon and thoron entering the second compartment are significantly different. The films in the two compartments thus permit simultaneous measurement of integrated radon and thoron concentrations. Fig.8



Radon dosimeter cup (Al).

The third film outside the cup is for the assessment of daughter product concentration.

The overall size of the dosimeter cup is suitable for its mounting on the waist belt of the uranium miners. The cup is shown in fig. 8.

11.3.3. Measurement of alpha activity in the effluents from various nuclear facilities

This particularly refers to releases of actinides which are long lived and are almost invariably alpha emitters. The intention of the investigations is to assess the impact of releases on the aquatic/marine food chain.

11.3.4. Neutron dosimetric measurements

This technique is playing an important role in biological dose measurements from neutrons in personnel dosimetry and is mainly based on recoil track registration. Improved availability of dosimetry grade CR-39 coupled with technique of electrochemical etching for films may soon put this method in an unenviable position.

11.4. Biological Applications

Applications here involve not only the conventional ones like dosimetry and particle identification but also exotic ones like microporous filters, described a little later.

11.4.1. Alpha activity content of blood

Alpha activity in the fresh blood samples is now being monitored

by immersing CR-39 detectors in it and freezing the whole arrangement subsequently. Smoking and non-smoking subjects are being compared this way.

11.4.2. Lead content of teeth and bone

SSNTD technique is being used to measure lead content and its distribution in teeth and bone and to relate it, if possible, with the age of the person. Radioactive isotopes ^{206}Po and ^{210}Po are produced from $^{206,207,208}\text{Pb}$ on bombardment with ^3He and ^4He ions. They have half lives of 8.8 days and 138 days respectively. Pulpal dentine (teeth) and Tibia (bone) sections show increase with age, giving clear indication that lead enters blood supply through food and inhalation of car exhaust and industrial pollution.

11.4.3. Filtration of malignant cells by microporous films

Charged particles (fission fragments and other heavy ions) have been used to produce uniform fine etched holes in thin sheets of mica and plastics. They are now commercially available with hole sizes in the range $0.03\mu\text{m}$ to $10\mu\text{m}$. They are being used to separate rigid large sized cells -cancerous or otherwise - from blood. These filters are therefore finding use as diagnostic tools in cancer studies and in the investigations of blood circulation disorders.

11.5. Track Detectors In Teaching

Until recently the availability of foil kits has made it possible to demonstrate certain basic nuclear phenomena using the activation foil technique. The simplicity and inexpensive nature of plastic track detectors however offers many advantages. SSNTD kits are now available which permit conduct of about a dozen experiments for under graduate/post graduate level. A list of simple experiments is given in Table 3.

Above all, the cheapness of the kit is clear proof that not all branches of modern science are expensive. This branch needs serious consideration at the level of Indian Universities. A number of references contain details of some of the above experiments; notably Bhagwat et al, 1976; Bhagwat, 1983; Bull, 1980a, 1980b, Enge, 1980; Durrani, 1982.

Table 3

A list of simple experiments based on SSNTD technique

-
1. Measurement of strength of radioactive sources by alpha/fission fragment track registration technique.
 2. Ranges of charged particles (alpha or fission fragments) in air and their variation with energy.
 3. Study of inverse square law for point sources.
 4. Determination of half life of short lived alpha emitters e.g. Cf-252 source.
 5. Specific activity measurement of long lived radio active materials like uranium/thorium etc.
 6. Ratio of alpha to fission decay for radioisotopes decaying by two modes, e.g. for Cf-252.
 7. Spontaneous fission decay constant of U-238.
 8. Determination of uranium content of glasses, minerals etc.
 9. Measurement of non-uniformity of deposition in electrodeposited/plancheted sources.
 10. Standardisation of optimum conditions of chemical etching for i) Lexan polycarbonate (for fission tracks) and ii) CN and CR-39 films (for alpha and fission tracks).
 11. Measurement of critical angle of track registration.
 12. Determination of activation energy of bulk etching.
-

11.6. Some Nuclear Applications

Prominent applications in nuclear industry include:

- i) Autoradiography of fuel pellets like (U-Pu) O₂, (U-Th)O₂ to study uniformity of mixing of two components and to evaluate concentration of one component in the other.
- ii) Measurement of low levels of alpha activity in a variety of matrices.
- iii) In-situ dosimetry of reactor components.
- iv) Preparation of track-etched microfilters.

11.7. Likely Future Applications of SSNTD

Although there can be numerous applications in this category, depending upon the ingenuity of the researcher, a few are mentioned below just to give a feel for the shape of things to be expected:

i) Development of practical neutron dosimetry systems based on electrochemical etching technique.

ii) SSNTD and electret combinations for environmental radon/thoron dosimetry.

iii) Charged particle radiography for biological and other applications.

iv) Studies involving interactions of charged particles less massive than protons (i.e. mesons) through development of more sensitive detectors like SR-86.

v) Search for slow moving supermassive magnetic monopoles (of mass greater than $10^{16} \text{ GeV}/c^2$).

12. CONCLUSION

In the end it may be added that the SSNTD technique, though of recent origin has carved a niche for itself in the field of radiation detection and measurements. There is no doubt that it will continue to score over other techniques by virtue of its sheer simplicity and low cost.

A small book of this kind can provide only a brief glimpse of the vast and growing subject, but we hope the readers are enthused to find new application of SSNTD in their own field.

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Dr. Bhagwat has also specialised in the field of Solid State Nuclear Track detection (SSNTD) and has been associated with it for about 2 decades now. He concentrates mainly on application of this technique to problems in health physics.

Dr. Bhagwat takes interest in teaching and has served on teaching faculty of BARC Training School for many years. He is a former editor of "Bulletin of Radiation Protection" and has more than 50 research papers to his credit.

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