CHAPTER - III

SOLID STATE NUCLEAR TRACK DETECTORS
3.1 Introduction

The field of solid state nuclear track detectors (SSNTDs) is about a quarter century old now. It was noticed that heavy charged particles produce submicroscopic tracks in many insulating solid materials including crystals, inorganic glasses and plastics (Fleischer et al., 1972). These submicroscopic tracks consist of radiation damaged materials produced by the passage of the heavy charged particles. The damaged regions are narrow and can be developed to suitable size for optical viewing under the microscope, they were first observed by Young of Atomic Energy Research Establishment, Harwell (Young, 1958). In the paper reported in 1958, the observation of fission fragment tracks in lithium fluoride (LiF), which are suitably etched with an etchant consisting of concentrated HF (1 part) and glacial acetic acid (1 part) saturated with ferric fluoride, were published.

The submicroscopic tracks can also be observed directly at a high magnification in an electron microscope as reported by Silk and Barnes (1959). They observed fission tracks in mica and concluded that the tracks were less than 300 Å in diameter and greater than 4 µm in depth. The above observations were part of the experiments conducted to study the nature of damage produced by radiation such as fission fragment in different materials.
The credit of development, from the above findings of a new particle detector with important advantages over earlier detectors in certain nuclear service applications, therefore, goes to Fleischer, Price and Walker of General Electric Company, Research and Development Centre, Schenectady (New York), who began their studies in 1961 from where Silk and Barnes had left. Their objective was to explore the utility of SSNTD in nuclear research and took for fossil tracks of cosmic ray induced events in meteorites.

Since then the field has grown to such an extent that there is hardly a branch of science and technology today where it does not have an application. Some of these fields are fission and nuclear physics, space physics, study of meteoritic and lunar samples, cosmic rays, particle accelerators and reactors, metallurgy, geology and archaeology, medicine and biology and others. Now-a-days the SSNTDs become popular in many international and national laboratories as well and the extensive use of the detectors are going on.

A list of commonly used solid state nuclear track detectors is given in the comprehensive book by Fleischer, Price and Walker (1975). Basically these are grouped as inorganic crystals, inorganic glasses and high polymers (plastics). The plastics constitute the most sensitive group and are commonly used in the study of heavy ions, cosmic rays, neutron dosimetry and radon/thoron dosimetry. Sensitivities of some commonly
used plastics are listed in Table - 3.1.

One of the most sensitive plastic is CR-39 (Allyl diglycol carbonate) which is able to register protons and relativistic nuclei with charges $\gtrsim 20$ (Cartwright et al., 1978). It has been successfully used for the relativistic cosmic ray iron nuclei (Biswas et al., 1979). Recently, it was reported that protons of energy up to $\sim 10$ MeV can also be revealed by etching in CR-39 plastics (Rao et al., 1982).

3.2. **Important Features of SSNTDs and Their Comparison with Other Detectors**

The density of ionization-damage produced by a moving particle in solids is directly proportional to the square of its charge and approximately inversely proportional to the square of its velocity. This very property of charged particles has been used very successfully by various workers in the study of cosmic ray particles through nuclear tracks.

It has been found, both theoretically and experimentally, that different materials have different sensitivities to nuclear particles. Organic polymers or plastics are found to be the most sensitive, some of which (Cellulose acetate, Cellulose nitrate, Cellulose acetate butyrate and CR-39) undergo detectable chemical changes when they are irradiated by low energy protons, neutrons and alpha particles etc. On
Table - 3.1

Relative Sensitivities of Some Plastics

<table>
<thead>
<tr>
<th>Plastic</th>
<th>Atomic structure</th>
<th>Least ionizing ion seen</th>
</tr>
</thead>
<tbody>
<tr>
<td>Amber</td>
<td>C₂H₃O₂</td>
<td>Full-energy fission</td>
</tr>
<tr>
<td>Polythene</td>
<td>CH₂</td>
<td>Fission fragment</td>
</tr>
<tr>
<td>Polyvinyl acetochloride</td>
<td>C₆H₉O₂Cl</td>
<td>42 MeV ³²S</td>
</tr>
<tr>
<td>Polymide</td>
<td>C₁₁H₄O₄N₂</td>
<td>36 MeV ¹⁶O</td>
</tr>
<tr>
<td>Bisphenol A-polycarbonate (Lexan, Makrofol)</td>
<td>C₁₆H₁₄O₃</td>
<td>0.3 MeV ⁴He</td>
</tr>
<tr>
<td>Polyoxyethylene (Delrin)</td>
<td>CH₂O</td>
<td>28 MeV ¹¹B</td>
</tr>
<tr>
<td>Polypropylene</td>
<td>CH₂</td>
<td>1 MeV ⁴He</td>
</tr>
<tr>
<td>Cellulose nitrate (Diacell)</td>
<td>C₆H₈O₉N₂</td>
<td>0.55 MeV ¹H</td>
</tr>
<tr>
<td>Cellulose nitrate (Kodak-Pathe)</td>
<td>C₆H₈O₉N₂</td>
<td>0.1 MeV ¹H</td>
</tr>
<tr>
<td>Allyl diglycol carbonate (CR-39)</td>
<td>C₁₂H₁₈O₇</td>
<td>10 MeV ¹H</td>
</tr>
</tbody>
</table>
the other hand, a few materials, e.g. some meteoritic minerals are so insensitive that they record only the passage of heavily damaging nuclei such as low energy iron ions. Heavy ion irradiation and experience with cosmic rays have shown that each track-detecting material has a well defined threshold damage density below which no tracks are produced. This threshold characteristic has contributed significantly to the establishment of the solid state nuclear track detectors as an important tool in many notable applications in the physical sciences.

The important features of solid state nuclear track detectors can be summarized as follows:

(i) The detectors are simple in construction and use;

(ii) They are insensitive to light (c.f. nuclear emulsion);

(iii) These detectors are very cheap and can be obtained in very small as well as very large sizes. Small detectors can be used for measuring the particle flux in very inconvenient locations while large detectors can be used to record very rare particle events;

(iv) A permanent record of particle tracks is obtained in them and can be left or stored for very long time unattended under severe environmental conditions like high temperature, pressure, humidity, radiation background and extreme mechanical vibrations;
(v) Heavy charged particles such as fission fragments can be recorded and distinguish from a very high background of lighter charged particles like $^4\text{He}$, $^1\text{H}$, $^2\text{H}$, beta-particles, X-rays and gamma rays (c.f. nuclear emulsions);

(vi) Due to the fact that the detectors can be placed in direct contact with the fission fragment sources, a very high efficiency and sensitivity can be obtained;

(vii) These detectors also possess the charge and energy discrimination properties. The resolution for high Z particles obtained with plastics has been reported to be better than that with nuclear emulsions;

(viii) The detectors have a considerable amount of geometric flexibility, and are therefore, particularly useful in angular distribution measurements;

On comparing the properties of SSNTDs with other detectors, it can be easily observed that SSNTDs have many advantages over other detectors in general, and nuclear emulsions in particular. The fact that they can be placed in direct contact with the fission sources and the fission fragment can be recorded and distinguish in a mixed field of unwanted light charged particles, neutrons and gamma rays, has made these detectors extremely useful in experiments where fission reaction rate is very low (Khan and Durrani, 1972) and a very high detection efficiency is required.
Light charged particles such as $^4\text{He}$, $^3\text{He}$, $^3\text{H}$, $^2\text{H}$ and $^1\text{H}$ were detected by the use of SSNTD and attempts were made to make some reasonable estimates of their energies. SSNTD was also applied in the study of (a) mass and energy distribution of fission fragments (Ait-Salem et al., 1963) and (b) blocking or channelling phenomenon in single crystals (Fleischer et al., 1975).

Despite these advantages, these detectors suffer from some disadvantages. The basic disadvantage with these detectors is that only a part of the total length of the particle trajectory can be seen and the part that has been etched off can not be recovered. Also very accurate energy determination is still not possible with these detectors.

3.3. **APPLICATIONS OF SSNTDs**

The technique of solid state nuclear track detection has found applications in almost every branch of science. The applications range from nuclear physics to biology, from the depths of the earth into the distant space, from the small to the large, and from ancient to the recent. Some of the main applications are given below:

3.3.1. **Nuclear Physics**

As the SSNTD system can perform free detection of heavy
charged particles (like fission fragments) in the presence of high dose of light charged particles (like p-particles etc.) and gamma rays and as they can be made insensitive to neutrons (if desired), they are ideal for fission studies. They have been employed advantageously in the determination of spontaneous fission decay constant of a number of heavy nuclides (Flerov et al., 1964).

Some of the other important applications of solid state nuclear track detectors in nuclear physics have been listed below:

(i) Ternary fission was first observed in 1963 (Fleischer et al., 1966).

(ii) Fission barrier and the saddle point mass of $^{201}$Tl was determined by Burnett et al. (1964).

(iii) Synthesis of element 104 was demonstrated (Flerov et al., 1965).

(iv) Flerov et al. (1964) measured the excitation functions and the isomeric yield ratio for the 14 ms fissioning isomer from deutron irradiation of plutonium.

(v) Determination of the isotopes of element 102 with mass 251 to 258 took place in 1966 (Flerov et al., 1966).

(vi) Angular an-isotropy and nuclear pair correlation effects in nuclear fission were observed by Smirenkin et al. (1968).
(vii) A number of track detectors were used in the investigations on the ternary photofission (Medveczky and Somogyi, 1970).

(viii) High energy photofission of heavy and medium heavy elements was achieved (Methasiri and Jahansson, 1971).

(ix) Determination of charge and energy of fragments emitted in relativistic heavy ion reactions were made.

(x) The determination of picobarn cross-section for the electrofission of $^{24}$Hg was reported by Chung in 1973 (Chung, 1973).

(xi) In 1975, some very energetic heavy fragments from relativistic heavy ion reactions observed by Crawford et al. (1975).

(xii) Some rare multiprong events were observed (Khan et al., 1980; Brandt et al., 1980).

3.3.2. Geology and Archaeology

Almost all materials are known to contain traces of natural uranium. Over the geological period, some of the uranium atoms decay through the spontaneous fission process, thus producing fission fragments. These fission fragments while flying apart create latent damage trails. The density of these 'natural' latent damage trails is given by

$$\rho = K.C. \text{ Age} \quad \text{... (3.1)}$$
where \( C \) = uranium contents of the sample;

\[ \text{Age} = \text{age of solidification of the sample}; \]

\[ K = \text{a constant, function of the geometry of the } \]
\[ \text{surface analysed, and the range of the fission } \]
\[ \text{fragments in the material.} \]

By exposing the same sample with thermal neutrons, and

counting the 'induced' track density, 'C' and hence 'K' are

estimated. Thus the 'Age' of solidification of the sample can

be determined (Fleischer et al., 1975). This method of dating

known 'Fission Track Dating' has been established over the

years and is now considered to be a standard technique with

certain unique advantages over the conventional methods. It

has been extensively used for the dating of geological, archaeo-

logical and cosmological samples. The method has been success-

fully applied in studies like Ocean Bottom Spread and the

Continental Drift (Fleischer et al., 1975).

The scanning of individual tracks enable to find not only

the contents but also the distribution of elements like Pu, U,

Th, B, Pb, Bi etc. in the sample of interest. This method was

extended to study the diffusion of the atoms of various ele-

ments in geological materials under varying environmental

conditions.

The method of 'Fission Track Dating' was applied success-

fully to date a number of archeological samples. The range of

applicability extended from the 'Stone Age' to the very recent
one. The track studies of 'Roman Glasses' and the 'Franchtic Cave' obsidians have yielded extremely interesting information regarding the Roman period and the history of Seafaring near the Greek Mainland (Durrani et al., 1971).

3.3.3. Cosmology

Plastic and glass track detectors have been employed in studying the cosmic ray fluxes at high altitudes of the earth and on the surface of the moon. The track studies of Apolo and Luna samples yielded important information concerning the past radiation and thermal histories, and the dynamic processes of lunar surface. These studies when coupled with microcrater work provided useful information about the composition and fluxes of micrometeoroid in space as a function of time (Smith et al., 1974).

The track studies of meteoritic objects made possible the measurement of the duration of various types of space exposures and provided information about the following processes:

(i) The erosion and accretion rates on the lunar surfaces;
(ii) The original size of meteorites prior to their loss of mass caused by ablation while entering the earth's atmosphere;
(iii) The time of fall of tektites on the earth;
(iv) The fluxes of cosmic ray particles existing during different periods in the past.
As recent in 1966, the known cosmic ray spectrum extended up to only iron. This 'Iron Curtain' was broken by Fleischer and co-worker in 1967, during their track studies of meteorites (Fleischer et al., 1967). They discovered many trans-iron nuclei present in cosmic rays.

Mcdougal and co-workers (1975), in track studies of meteorites, observed that the outer surfaces of the individual grains of certain meteorites indicated exposure to cosmic rays. Further work on these grains indicated that the grains enjoyed independent existence before joining to form bigger meteoritic bodies. The fission track analysis and cosmic ray exposures yielded information about the dates of their independent existence, and their later thermal and radiation histories.

3.3.4. Biology

If a thin plastic track detector (previously bombarded with charged particles) is etched, 'through holes' are produced. The size, shape, number and the position of these holes can be controlled by the exposure and etching conditions. Detectors containing such through holes have been used in filtering of cancer cells from human blood. They have also been employed for cleaning air from dust particles and other suspensions.

Track detectors have also been used in the radiobiology of plutonium. The importance of these studies lies in the
fact that due to extensive involvement in plutonium production, the danger of its intake by the working personnel has increased. Track detectors have been employed for mapping the locations of plutonium concentrations in living matter. There are two methods of getting the required plutonium maps. First, the detector is put in contact with the tissue and then the direct autoradiograph of natural alpha decay is obtained. A second more rapid mapping is achieved by irradiation of the 'detectors-tissue' assembly by thermal neutrons. The resulting fission fragments yielded the required mapping. The results so obtained have shown that plutonium concentrates on bone surface and in particles within the marrow. Such studies have been made on rabbits, beagles, rats, and on human subjects. An image resolution of about ten microns has been achieved in these experiments.

3.3.5. Ancient Nuclear Reactors

Some years ago, ancient natural nuclear reactor sites were discovered in Oklo (Gabon). Track analysis of crystalline materials collected from various locations yielded important information regarding (a) the reactor side, (b) the time period elapsed after the activity stopped, (c) the total time for which the reactors remained active, (d) estimates of the power levels of the reactors, (e) burn up etc.
Further analysis is expected to yield still more interesting information regarding these ancient natural reactor systems.

3.3.6. Bird Altimetry

An interesting application of alpha sensitive plastic track detectors was made by Kristeansson et al. (1977). They built a simple integrating barometer and employed it for the measurement of the distribution of flight altitudes of birds (Kristeansson et al., 1977). They fixed an alpha source near a sensitive detector (the total weight less than one grain) so that the alpha particles could reach only the nearest position of the detectors, while the 'barometer' was on the surface of the earth. When the birds took the 'barometer' to higher altitudes, due to lower atmospheric density, the ranges of alpha particles increased. This caused the track formation further along the detector length. A study of the distribution of the ranges of tracks along the detector length, yielded interesting information about the distribution of the time spent by the birds at different altitudes. The results of these experiments provided the favorite altitudes of a variety of birds both in dry and wet weathers.
3.4. CR-39, A NEW ALPHA SENSITIVE PLASTIC TRACK DETECTOR

In the recent past various types of dielectric detectors have become firmly established as more economic alternatives for the radiation dosimetry. LR-115 has been found to be quite vulnerable to environmental conditions, while the manufacturers of CA 80-15 (Kodak Pathe, France) discontinued its further fabrication and, instead, have introduced a new product named CN-85. Recently a new type of plastic track detector was reported (Cartwright et al., 1978; Cassau and Benton, 1978). This new type of detector has been named as CR-39 (manufactured by Pershore Mouldings Ltd., Pershore, U.K.) and was reported to have some unique characteristics (Al-Najjar et al., 1979).

Most plastic detectors suffer the physical disadvantages of inhomogeneity and anisotropy, after chemical etching these features manifest themselves in many ways including unevenness of track profiles, inhomogeneities of bulk etching rate giving rise to random surface pitting and different response characteristics to etching between surfaces of the same sheet, together with other factors related to the amounts and distributions of plasticizers and stabilizers within an individual sheet of plastic. CR-39 polymer is highly cross-linked thermoset prepared by polymerisation of diethylene glycol bis (allyl carbonate) monomer. The polymerisation is initiated by radicals and proceeds via the addition reaction of allyl
groups. The structure of the resulting polymer is a dense three-dimensional network consisting of polyallyl chains joined by diethylene glycol dicarbonate links. The average length of polyallyl chain is relatively short ($DP_w \approx 80$) as the result of degradative chain transfer specific for polymerisation of allyl groups. The formation of a network during polymerisation is a special case of the percolation process and can be modelled by a Monte Carlo simulation. The simulation predicts the distribution of cluster size, the gel point and the length distribution of the polyallyl chains as a function of monomer conversion. It can predict inhomogeneities in the cross-linking density which could explain some anomalous etching behaviour of CR-39 (Stejny, 1987). The polymerisation does not reach full conversion because the system enters the glassy state first. However, the residual mobility in the glassy state and the presence of chemically reactive group are responsible for physico-chemical aging of the polymer and the changes in the performance of CR-39 with time. The molecular structure of CH-39 polymer is given below:

```
O
\CH_2 - CH_2 - O - C - O - CH_2 - CH = CH_2
/\
O
\CH_2 - CH_2 - O - C - O - CH_2 - CH = CH_2
/\O
```
CH-39 has an abrasion resistance similar to glass, a specific gravity of 1.32 (25°C), a refractive index of 1.505 (20°C) and is isotropic and homogeneous with respect to its ability to record ionizing radiation from alpha particles. Cross-linking after radiation damage of chemical bonds does not occur, hence effective track resolution is enhanced compared with many other plastic detectors. It is resistant to most solvents except strong bases and highly oxidizing acids (Hamilton and Clifton, 1981). These are amongst the advantages that make it superior to most other plastics commonly used for nuclear track detection. Reproducibility of track production in the detector over a range of curing times and etching temperatures was excellent as indicated in Table - 3.2 (Clifton and Hamilton, 1981), in which measurements of the area of tracks at 90° incidence to the detector were recorded after irradiation of CR-39 with 5.5 MeV α-particles from 241Am.

Comparing to other plastic detectors, CR-39 is extremely sensitive to ionizing radiation having a minimum detectable ionization, \( Z/\beta \approx 18 \) compared to \( \approx 30 \) for cellulose nitrate. In Table - 3.3 approximate \( E_{\text{min}} \) and \( E_{\text{max}} \) values in some plastic detectors are presented. CR-39 possesses a very high degree of isotropy and homogeneity and it records tracks of low energy alpha particles easily. The clarity of track recorded is unsurpassed and the signal-to-noise ratio unprecedented in CR-39 than cellulose nitrate and polycarbonate plastics.
<table>
<thead>
<tr>
<th>Etching time (m)</th>
<th>Temperature (°C)</th>
<th>Track area (µm) for α-particles recorded at an angle of 90° in CR-39 for different curing times (h)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>8h</td>
</tr>
<tr>
<td>120</td>
<td>68.0</td>
<td>&lt;5</td>
</tr>
<tr>
<td>230</td>
<td>74.0</td>
<td>42.7±3.8</td>
</tr>
<tr>
<td>420</td>
<td>72.0</td>
<td>118.5±8.0</td>
</tr>
<tr>
<td>120</td>
<td>82.0</td>
<td>34.7±2.5</td>
</tr>
<tr>
<td>400</td>
<td>82.0</td>
<td>376.2±4.5</td>
</tr>
</tbody>
</table>
Table - 3.3

Limits of Detectable Alpha Particle Energies for Some Plastics

<table>
<thead>
<tr>
<th>Type of plastic</th>
<th>$E_{\text{min}}$ (MeV)</th>
<th>$E_{\text{max}}$ (MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lexan Polycarbonate</td>
<td>~0.2</td>
<td>~0.4</td>
</tr>
<tr>
<td>Makrofol Polycarbonate</td>
<td></td>
<td></td>
</tr>
<tr>
<td>CA80-15 Cellulose Nitrate</td>
<td>~0.1</td>
<td>4-6</td>
</tr>
<tr>
<td>CN-85 Cellulose Nitrate</td>
<td></td>
<td></td>
</tr>
<tr>
<td>LR-115 Cellulose Nitrate</td>
<td></td>
<td></td>
</tr>
<tr>
<td>Daicel Cellulose Nitrate</td>
<td></td>
<td></td>
</tr>
<tr>
<td>CR-39 Allyl Diglycol Carbonate</td>
<td>~0.1</td>
<td>&gt;50</td>
</tr>
</tbody>
</table>
Exposures to a variety of gamma and beta particles (0.16 - 2.8 MeV) emitters for 1 h at a dose rate of 10^6 R/h at contact, no interference or impairment to the surface of the detector was observed. The effect of UV radiation on the damage tracks of alpha particles and fission fragments in CR-39 and lexan was reported by Wong and Hoberg (1982). They reported a 30% increase in the track density caused by UV exposures while the track diameter is enhanced by 100%. The \( V_T/V_B \) and hence the efficiency remains nearly constant for CR-39 plastic.

A number of investigators have measured the etching characteristics of CR-39 plastics under various concentrations of NaOH and KOH (Green et al., 1982; Amin and Henshaw, 1981). These studies show that 6N NaOH at 70°C represents the optimum etching conditions for CR-39 (Green et al., 1982). The etching characteristics of CR-39 detectors measured by the author for radon monitoring show the same optimum conditions as reported by Green et al. (1982), details are given elsewhere (Khan, 1986).

It is established that the CR-39 plastic detector is the most sensitive detector among all available detector and thus applied in almost every branch of science and technology (Henshaw et al., 1981). CR-39 plastic track detector has proved the best for fast neutron dosimetry (Bradley et al., 1986; Spurny et al., 1987; Pitt and Werner, 1988).
The results obtained so far strongly suggest that CR-39 is not only the most sensitive track detector existing today for the detection of alpha particles from radon and thoron, but that it also has some unique properties which make it an ideal track detector for radon/thoron measurement (Khan and Ahmad, 1981) and in alpha autoradiography (Henshaw et al., 1979; Fews and Henshaw, 1984). CR-39 plastic detector is capable of detecting highly relativistic nuclei of the iron group and of higher charges in the cosmic radiation (Biswas et al., 1979).

Some of the useful properties of CR-39 are mentioned below:

(i) Among the plastic detectors, CR-39 plastic have maximum track registration efficiency.

(ii) The track etch velocity for a certain heavy charged particles is the maximum in CR-39 plastic (Khan and Khan, 1983).

(iii) Fission fragment and alpha particle latent damage trails on CR-39 are highly stable (almost negligible annealing at temperature as high as 200°C) (Khan and Ahmad, 1981).

(iv) At present, CR-39 is the only plastic track detector which exhibits a combined linear and supralinear response (Luck, 1982).
(v) CR-39, is the most sensitive track detector at present. It registers tracks of intermediate energy ions of $^{18}O$, $^{16}O$ and $^{12}C$. Its efficiency for even light charged particles, such as protons and alpha particles having energy $E \leq 10$ MeV/n, is the highest among the presently known plastic track detectors (Khan and Khan, 1983).

3.5. FORMATION OF PARTICLE TRACKS IN SOLIDS

It is well known that a charged particle passing through an electrical insulator, produces a narrow region of radiation damaged material, known as 'Latent Track' or simply as a 'Track'. There are many theories given by the workers in the field to explain the mechanism of primary track formation. 'The Thermal Spike Model' is the first one in this direction (Bonfiglioli et al., 1961). The model shows that the charged particles passing through the detector solid produces local heating and there is a phase change causing dislocations in the material. The 'Thermal Spike Model' could not be well established for the track formation because the experiments did not show definite correlations between the sensitivities of the detector materials and temperature of phase changes.

It was Fleischer, Price and Walker (1965) who gave the so called 'Ion Explosion Spike Model' which has successfully explained the track formation mechanism. According to this
model, a positively charged particle knocks out the orbital electrons of atom lying in and around the vicinity of its path, thus producing a cylindrical region full of positive ions. These positive ions thereupon repel one another, disturbing and distorting the regular lattice in a crystalline solid (Figure 3.1) and producing more or less cylindrical region. On the other hand, in an organic polymer, the charged particle breaks the long molecular chains by ionization and excitation (Figure 3.2) and new species which are highly chemically reactive are formed. These latent damage trails can be seen at very high magnifications (30,000 X) under a transmission electron microscope. However, the electron microscope is not suitable for the observation of these latent tracks for various reasons. These difficulties may be overcome by the use of an 'etching process', i.e., the damage trails can be 'developed' and 'fixed' by using an appropriate etchant (Young, 1958).

Based on this model, the requirement for the track formation is that the Coulomb repulsive forces within the ionized region should be sufficient to overcome the lattice bonding forces, i.e., the electrostatic stress should be greater than the mechanical strength or bonding strength.

If two ions in a material of dielectric constant \( \varepsilon \) and average atomic spacing \( a_0 \) have received an average ionization of \( n \) unit charges \( e \), the Coulomb forces between them is
Figure 3.1. Track formation in a simple crystalline solid.

Figure 3.2. The Atomic Character of a particle track in polymer.
$n^2e^2/\varepsilon a_0^2$ and the force per unit area or the electrostatic stress will be $n^2e^2/\varepsilon a_0^4$. If $Y$ is the Young's modulus of the detector material, the bonding force or the mechanical strength in the lattice is given by $Y/10$. Hence according to the model, the first condition for the track formation is that

$$\frac{n^2e^2}{\varepsilon a_0^4} > \frac{Y}{10}$$

or

$$n^2 > R \equiv \frac{Y\varepsilon a_0^4}{10e^2} \quad \ldots \quad (3.2)$$

where $R$, defined in equation (3.2), is called the 'Stress Ratio' and is a measure of the relative sensitivity of various materials in which the tracks are formed. Thus the substance having smaller values of $R$, i.e., smaller value of mechanical strength, dielectric constant and lattice distance, should be more sensitive. The calculated values for stress ratio in some SSNTD are given in Table - 3.4. It is understandable that the plastics are more sensitive than inorganic glasses which in turn are more sensitive than inorganic crystals. This conclusion is in complete agreement with the experimental results (Fleischer et al., 1965).

3.6. TRACK REGISTRATION CRITERIA FOR SSNTD

In SSNTD, the track of heavily ionizing particles can be
Table - 3.4

Detector Materials in Order of Increasing Sensitivities

<table>
<thead>
<tr>
<th>Detector Material</th>
<th>Atomic number of lightest detectable charged particle</th>
<th>Stress ratio (R)</th>
<th>Composition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Olivine</td>
<td>23</td>
<td>1.7</td>
<td>MgFeSiO₄</td>
</tr>
<tr>
<td>Zircon</td>
<td></td>
<td>1.3</td>
<td>ZrSiO₄</td>
</tr>
<tr>
<td>Laboradorite</td>
<td>20</td>
<td>1.2</td>
<td>Na₂Ca₃Al₈Si₁₂O₄₀</td>
</tr>
<tr>
<td>Hypersthene</td>
<td></td>
<td>1.2</td>
<td>Mg₁.₅Fe₀.₅Si₂O₆</td>
</tr>
<tr>
<td>Phosphate glass</td>
<td>16</td>
<td>0.4</td>
<td>6₃P₂O₅:11UO₂:8Al₂O₃:9Ag₂O:9K₂O</td>
</tr>
<tr>
<td>Soda-lime glass</td>
<td></td>
<td>0.4</td>
<td>6₇SiO₂:14Na₂O:14CaO:5Al₂O₃</td>
</tr>
<tr>
<td>Orthoclase</td>
<td>15</td>
<td>0.4</td>
<td>KA₃Si₃O₈</td>
</tr>
<tr>
<td>Quartz</td>
<td></td>
<td>0.5</td>
<td>SiO₂</td>
</tr>
<tr>
<td>Amber mica</td>
<td>13</td>
<td>0.06</td>
<td>KA₃Si₃O₁₀(OH)₂</td>
</tr>
<tr>
<td>Potash mica</td>
<td></td>
<td>0.03</td>
<td>KMg₂Al₂Si₃O₁₀(OH)₂</td>
</tr>
<tr>
<td>Lexan polycarbonate</td>
<td>2</td>
<td>0.008</td>
<td>C₁₆H₁₄O₃</td>
</tr>
<tr>
<td>Cellulose acetate</td>
<td>1</td>
<td>0.009</td>
<td>C₁₂H₁₆O₈</td>
</tr>
<tr>
<td>Cellulose nitrate</td>
<td></td>
<td>0.02</td>
<td>C₆H₇O₁₁N₃</td>
</tr>
</tbody>
</table>
revealed by a suitable chemical etching. The usefulness of these detectors depends primarily on the ability to register heavy particle tracks without being affected by a dense background of less highly ionizing radiation that does not form tracks (Fleischer et al., 1965).

In the earlier experiments (Price and Walker, 1962a; Price and Walker, 1962b), it was noticed that in SSNTD, certain particles will be able to produce an etchable tracks in a certain solid while others do not, others may produce in another solid and so on. For example, the alpha particle can produce an etchable track in cellulose nitrate, cellulose acetate and polycarbonate plastics but do not produce tracks in polythene plastics like melinex, hostaphan, mayler and terphan, but it was noticed that the fission fragments were able to register tracks in these plastics. It has also been observed that the mica, is good for registration of fission fragments and not for light particles.

3.6.1. Total Energy Loss Rate Model

Fleischer et al. (1964) suggested that in the solids, only those particle will register the tracks which have a total rate of energy loss of an ion greater than a threshold value of \( \frac{dE}{dx} \) i.e. \( \frac{dE}{dx} \) \(_\text{crit} \), required by that solid. This \( \frac{dE}{dx} \) \(_\text{crit} \) was considered to be the characteristic of the solid.
They calculated the values of \( (dE/dx) \) and plotted as a function of energy per nucleon for various heavy ions (Ne, Si, S, Cl, Ar) in three solid detectors muscovite mica, lexan polycarbonate resin and cellulose nitrate. The data were consistent with the hypothesis that for each solid there exists a critical rate of energy loss \( (dE/dx)_{\text{crit}} \) such that particles losing energy more rapidly than this value produce continuous tracks with unit efficiency, while those depositing appreciably less energy per unit length produce no tracks.

Energy-loss curves for muscovite mica for the ions of Ar, Cl, S, Si and Ne between 0.35 and 10 MeV/n are shown in Figure 3.3. On these curves they displayed their observed data for long tracks, partial tracks and no tracks. It can be seen from the observations that in all the cases (for muscovite mica, lexan polycarbonate and cellulose nitrate), a transition region existed, above which the sensitivity of the detector is unity and below it is zero. In the case of transition region, also a partial registration of tracks can be observed. It can also be predicted that in cellulose nitrate plastic, the relativistic iron nuclei could be registered.

3.6.2. Primary Ionisation Criterion for Track Registration

Some new measurements on track registration in solids
Figure 3.3. Shows the $(dE/dx)_{\text{crit}}$ as the track registration criterion.
made by Fleischer et al. (1967) suggest that the results are inconsistent with $dE/dx$ criterion. The results show that the tracks will be registered in all three solids (muscovite mica, lexan polycarbonate and cellulose nitrate) at energy loss rates which are below the predicted threshold. Further, it was incorrect prediction that the relativistic iron nuclei should form tracks in cellulose nitrate.

For this purpose they adopted the so called 'Ion Explosion Spike Model' (Fleischer et al., 1965) for track registration in the dielectric solids. This model predicts that a quantity somewhat different from $dE/dx$ should determine the presence or absence of tracks viz. the number of ions formed per unit distance along the particle path. This quantity, called $dJ/dx$, is the primary specific ionization, given by Bethe (1930) as:

$$
\frac{dJ}{dx} = \left( \frac{\alpha Z e^2}{I_o \beta^2} \right) \left[ \ln \left( \frac{2 m c^2 \beta^2}{(1-\beta^2)I_o} \right) - \beta^2 + 3.04 \right]
$$

(3.3)

where $Ze$ is the effective charge of the ionizing particle, $\beta$ is the ratio of its velocity to $c$, the velocity of light, $m$ is the electron mass, $I_o$ is the ionization energy of the outer electron of the material, and $\alpha$ is the constant that depends on the material.

According to this criterion an etchable track in a solid will be formed if the linear ion density produced by the primary particle along the trajectory is greater than a critical value
required by the material. It also explains that a solid would register a track if the rate of primary ionization of \( \frac{dJ}{dx} \) was greater than a critical rate of primary ionization i.e. \( (dJ/dx)_{\text{crit}} \), which is a characteristic of the material.

Fleischer et al. (1967) plotted the value of primary ionization for different ions using the above equation in cellulose nitrate. From the curve 3.4, one can conclude that for each solid it is possible to choose a critical value of the primary specific ionization above which tracks are formed and below which there is no preferential etching of the solid.

It is important to note that not all ionization or bond breaking is included in equation 3.3. The secondary ionization produced by delta rays generally occurs to the side of the main particle track and, according to the ion explosion spike model, is largely irrelevant to its formation. This view is opposite to the view of Pfohl et al. (1965) and Benton et al. (1969), which is that the low energy delta rays play a vital role in the mechanism of track formation in polymers. In fact, the major difference between the primary ionization and the energy loss criteria is due to the increasing energy carried by the ejected electrons at higher energies of the ionizing particles. From the ion-explosion spike model they expect that only the number and not the energy of the removed electron is of importance as long as the electrons receive enough energy to drive them out of the track region.
Figure 3.4. Damage density (or ionization rate) vs velocity.
There are some objections to critical primary ionization criterion of Fleischer et al., which are:

(i) The values of dimensionless constants of stopping materials as used by Fleischer et al. are strictly true for hydrogen ($I_0 = 13.6 \text{ eV}$). Thus the absolute values of $(dJ/dx)_{\text{crit}}$ are not known.

(ii) The effect of delta rays or high order of ionization has been neglected.

(iii) The value of $I_0 = 2 \text{ eV}$ utilized by Fleischer et al. for plastics appears to be inconsistent with the ion explosion spike mechanism which requires electron ejection from the track region. The energy of 2 eV may be sufficient to break a chemical bond (raise the bound electron to a higher energy states) but in general it is not sufficient to ionize the atom. For ionization a higher value of 9-15 eV is needed (Bovey, 1958). Also the optical absorption measurements indicate that lexan is highly transparent to 2 eV photon ($\lambda = 6200 \text{ A.U.}$).

3.6.3. Critical Restricted Energy Loss Rate Criterion

In view of the objections to primary ionization criterion, Benton and Mix (1969) proposed a new track formation criterion for plastic track detectors, known as 'Restricted Energy Loss' (REL) criterion. For this purpose, they incorporated the
secondary ionization and excitations produced by the low energy recoil electrons (low-energy delta rays). They assumed that the total energy deposited per unit track volume by the incident charged particles determines the chemical reactivity of the latent track region.

It has been established that the relativistic charged particles of high energy can produce delta rays (having the energy of several MeV) along the particle trajectory and these delta rays tend to be scattered and deposit their energy at a considerable distance from the path of heavy ion. Benton argues that the plastics are generally 100 μm thick, the energetic delta rays and most of their accompanying energy are lost from the detector and so this energy is not considered in track formation. Hence, the total rate of energy loss (dE/dx) of a charged particle can not be used as criterion for track registration.

According to restricted energy loss rate criterion, the secondary electrons having the energy ω less than a predetermined value ω₀, in addition to the primary ionization, contributed in the track formation. If the energy density is greater than the minimum value depending upon the nature of the detector, a particle will form a track.

Benton and Nix (1969) calculated (dE/dx) ω < ω₀ for different particles using the following relations:
\[
\left( \frac{dE}{dx} \right)_{\omega < \omega_o} = \left[ 2\pi n(Ze)^2 r_o^2 m_o c^2 / \beta^2 \right] \left[ \ln \left( 2 m_o c^2 y^2 \omega_o \right) / I_{adj} \right] \\
- \beta^2 - 2(c/Z) - \delta \] 
\]

... (3.4)

where \( Ze \) = Effective charge of the ionizing particle,
\( n \) = Density of electrons in the stopping material,
\( r_o = e^2 / (m_o c^2) \), the classical electron radius,
\( y = (1 - \beta^2)^{-1/2} \),
\( I_{adj} \) = Mean excitation potential of the material,
\( c/Z \) = Tight binding shell correction
and \( \delta \) = correction for the density effect.

The effective charge, \( Ze \), is calculated by the equation
\[
Ze = Z \left[ 1 - \exp \left( -130 \, \beta / Z^{2/3} \right) \right] \] 
\]

... (3.5)

where \( Z \) is the nuclear charge of the particle. If the restricted energy loss (REL) rate, given by equation 3.4, is greater than a minimum value \( [(dE/dx)_{\omega < \omega_o}]_{crit} \), depending upon the detector, the tracks are formed. Thus according to this model, a particle will produce an etchable track where \( (dE/dx)_{\omega < \omega_o} \) will be greater than \( [(dE/dx)_{\omega < \omega_o}]_{crit} \), i.e.,
\( (REL) > (REL)_{crit} \) for the material. It must be noted here that while the primary ionization model does not give the value of \( (dJ/dx)_{crit} \) in absolute units, the restricted energy loss model gives the value of \( [(dE/dx)_{\omega < \omega_o}]_{crit} \) in absolute units.
Benton concludes that ion explosion is not essential for track formation in polymers. What is required is the production of a sufficient number and density of chemically reactive species. These species may be formed by low energy delta rays. The distribution of these chemically reactive species near particle trajectory is unknown. It is reasonable to assume that a minimum concentration of these centres is needed in order to be able to develop a track through the chemical etching techniques.

3.6.4. Critical Dose of Ionization Energy Criterion (D_{crit})

Katz and Kobetich (1968) reported a new criterion for track formation in solid state nuclear track detectors. They argued that at higher energies the principal mode of energy loss is through ionization, the tracks are formed through the deposition of energy of secondary electrons or delta rays in the immediate vicinity of the particle's trajectory. They suggested that a track in detector will be produced when a critical dose, (D_{crit}), of ionization energy is deposited by the secondary electrons at a critical distance, (R_{crit}), from the particle's path (\sim 20 \text{ Å}).

The spatial distribution of ionization energy in the vicinity of the path of the incident particle by combining the delta rays energy spectra and the range-energy relation
of these electrons is calculated (Kobetich and Katz, 1968). They plotted these as a function of particle energy for different particles in mica, cellulose nitrate and lexan polycarbonate plastics. From these curves, it was found that there exists a critical dose, \( D_{\text{crit}} \), to be deposited at a critical distance, \( R_{\text{crit}} \), from the particles path, above which the tracks are formed and below which no tracks will be formed.

Although this criterion can also be fitted to most of the experimental data, it is difficult to evaluate this approach in detail because the results strongly depend upon the validity of basic assumptions: (a) that the contribution of the 'primary excitation' can be completely neglected, (b) the exact form of range-energy relations for electrons in eV region and (c) other assumptions in the calculations such as the form of angular distribution of the ejected electrons.

3.7. METHODS OF TRACKS VISUALIZATION

3.7.1. By Using Electron Microscope

Tracks produced by heavily multicharged particles such as fission fragments can be seen directly by an electron microscope in certain crystalline substances such as mica (Silk and Barnes, 1959). Electron microscope has commonly been used in two modes.
3.7.1.1. **Differaction contrast mode**

In this method the crystal planes of the detector are bent such that electrons get scattered out of the Bragg reflection image of the damaged regions which appear as black streaks. Due to the bending the strain is distributed around the tracks and hence one gets the upper limit of the damaged region. The diameter of tracks in the irradiated mica was found to be 100-150 Å (Silk and Barnes, 1959).

3.7.1.2. **Thickness contrast mode**

The irradiated thick sheet of a detector is viewed under an electron microscope after the preliminary etching. The image appears as light or transparent line. Price and Walker (1962) observed the tracks in the irradiated etched mica using this method.

However, electron microscope method has following limitations:

(a) extremely thin (< 3000 Å) samples are required,
(b) large densities of tracks are needed,
(c) only small track lengths can be examined,
(d) tracks of only very heavy charged particles can be seen.

3.7.2. **Track Decoration or Precipitation Method**

The most successful application of this method has been
made by Childs and Slifkin (1962a; 1962b; 1963), who have decorated tracks throughout the volume of a thick single crystal of Silver Chloride. They used a combination of pulsed light and electric fields to sweep photoelectrons into the interior of the damaged part of the crystal. Multipronged events from 1.5 GeV proton interactions, heavy cosmic ray primaries and polonium alpha-particles tracks have been observed by this method. Although relativistic protons and alpha-particles do not give developable tracks, relativistic C, N and O groups register with unit efficiency.

An application of this precipitation method to decorate the tracks in a glass doped with silver was reported by Fleischer et al. (1963) using adequate heat treatment and electric field along the particle trajectory but no further work has been done using this method.

3.7.3. Chemical Etching Method

This is the most commonly and widely used method to reveal the tracks in solid state nuclear track detectors. The method was first used by Young (1958) for revealing the fission fragment etch pits in thick sample of Lithium Fluoride (LiF). The method is based on the following principles:

As the damage trails are highly reactive regions (compared to the undamaged part), on immersing the sample in an appropriate etchant, all those trails which intersect the surface
are rapidly dissolved and hollow cylindrical tubes about 50 Å in diameter are left behind (Enge, 1980). If the procedure is prolonged, the etching continues at slightly lower rate. The undamaged wall of the hollow tube is also attacked and its diameter increases into micron range (which is comparable to the wavelength of visible light), thus ultimately, a modified, enlarged version of the original damage trail is produced, which can be seen easily under an optical microscope (Khan, 1983).

The shape of an etched track in a certain material depends not only on the charge, mass and velocity of the incoming particle and on the nature, concentration and temperature of the etchant but also on the environmental conditions at the time of irradiation and pre-etching treatments. One of the important parameters governing the shape of the track is the critical angle of etching (the angle below which particles falling on the detector surface will not be revealed by etching) of the material for a particular particle of a specified energy determined under specified experimental conditions. In lexan, makrofol, mica etc., for beams of fission fragments of normal energy, the resulting enlarged tracks are long and almost cylindrical with slightly tapered ends (Khan and Durrani, 1972). Charged particles produce conical etch-pits in glasses (Khan and Durrani, 1972) which appear circular or oval when viewed from above. In the past, choosing the best etchant has largely
been a matter of trial and error, which is not so now. Work has been done in this direction from last over fifteen years and some guidelines have been developed. In Table - 3.5, etching conditions of some such detectors are given.

The geometry of a track formed by a particle is governed by simultaneous actions of the etchant on the bulk of plastic and along the damaged region produced by the particle in the detector material. When the irradiated detector is immersed in a suitable chemical reagent under pre-determined conditions, the etchant dissolves the bulk material at the rate of $V_B$-bulk etching rate, while its rate along the damaged region is represented by $V_T$-track etch rate. The damaged region gets dissolved faster than the bulk medium i.e., $V_T > V_B$. If the relative damage is measured in terms of primary ionization, $J$. Then of $J > J_c$, $V_T > V_B$, and $J = J_c$, $V_T = V_B$. The track registration geometry is given in Figure 3.5.

Different stages of chemical etching of a particle track in a SSNTD are shown in Figure 3.6. A charged particle enters in a detector and stops after traversing a certain depth (Figure 3.6a). The detector is immersed in an etchant under pre-determined conditions. If the final track is formed in time $t_0 = l_o/V_T$, where $l_o$ is the track length, layer of thickness '$V_B\cdot t_0$' will also be removed from the detector surface during the same time $t_0$, as shown in Figure 3.6b. Finally one can get a conical track (Figure 3.6c) of maximum length $l_m$. 

### Table - 3.5

**Etching Conditions of Track Etch Detectors**

<table>
<thead>
<tr>
<th>S.No.</th>
<th>Type of detector</th>
<th>Etching condition</th>
</tr>
</thead>
<tbody>
<tr>
<td>1.</td>
<td>Soda Lime Glass</td>
<td>48% HF, 22°C</td>
</tr>
<tr>
<td>2.</td>
<td>Vitreous Quartz Glass</td>
<td>48% HF, 22°C</td>
</tr>
<tr>
<td>3.</td>
<td>Phosphate Glass</td>
<td>10N NaOH, 50 ± 1°C</td>
</tr>
<tr>
<td>4.</td>
<td>Muscovite Mica</td>
<td>48% HF, 22°C</td>
</tr>
<tr>
<td>5.</td>
<td>Makrofol-N</td>
<td>6N NaOH, 50 ± 1°C</td>
</tr>
<tr>
<td>6.</td>
<td>Hostaphan</td>
<td>33% 6N NaOH + 33% H₂O + 33% CH₃OH, 40 ± 1°C</td>
</tr>
<tr>
<td>7.</td>
<td>Makrofol-E</td>
<td>6N NaOH, 50 ± 1°C</td>
</tr>
<tr>
<td>8.</td>
<td>Lexan</td>
<td>6N NaOH, 50 ± 1°C</td>
</tr>
<tr>
<td>9.</td>
<td>LR-115</td>
<td>6N NaOH, 60 ± 1°C</td>
</tr>
<tr>
<td>10.</td>
<td>CR-39</td>
<td>6N NaOH, 70 ± 1°C</td>
</tr>
</tbody>
</table>
Figure 3.2. Track registration geometry.
Figure 3.6. Principle of track etching.
given by:

\[ l_m = l_0 \left(1 - \frac{V_B}{V_I \sin \delta}\right) \ldots \quad (3.6) \]

where \( \delta \) is the dip angle of the particle, measured from the detector surface. The half cone angle of this conical track is given by

\[ \theta = \sin^{-1} \left(\frac{V_B}{V_I}\right) \ldots \quad (3.7) \]

Further etching causes the apex to become rounded (Figure 3.6d). When the particle penetrates the entire thickness of the detector material (Figure 3.6e) etching proceeds from both the sides, resulting in two sharp tipped cones known as double cone (Figure 3.6f). If the etching is allowed to continue long enough, the two cone merge and produce a hole, called etched through track (Figure 3.6g).

It should be noted here that not all the particle tracks which intersect the detector surface can be developed through this method of chemical etching. As already pointed out, in order to produce an etchable track, a charged particle must produce a damage density (i.e., rate of ionization) greater than a certain critical value which is the characteristic of the material. The quantity \( \frac{V_I}{V_B} \) is a function of the rate of ionization of the charged particle. It increases as the rate of ionization rises. For light ionizing particles,
$V_T/V_B \to 1$ and tracks are not observed. There is also a geometric limitation for getting observable tracks in these detectors. According to this limitation, tracks of those particles which enter the detector surface at an angle less than a critical angle given by $\theta_c = \sin^{-1} \left( \frac{V_B}{V_T} \right)$, can not be observed. In this case, the surface dissolves more quickly than the track develops (Somogyi, 1980). This can be obtained by putting $l_m = 0$ in equation and representing $\delta$ as $\theta_c$, as follows:

$$0 = l_0 \left( 1 - \frac{V_B}{V_T} \sin \theta_c \right)$$

or

$$\theta_c = \sin^{-1} \left( \frac{V_B}{V_T} \right). \quad \ldots \quad (3.8)$$

Thus, it is desirable to have the etch rate ratio as large as possible.

The bulk etching rate ($V_B$) depends on many factors like composition of the detector material, manner of the sample preparation, type of the etchant, etching conditions etc. Similarly the track etching rate ($V_T$) is also a function of the type and velocity of the incident particle. Hence $V_B$ provides information about the chemistry of the sample and about the etching process, while $V_T$ provides information about the charged particles.
Due to the existence of critical angle, the detection efficiency of the detector can not be equal to unity. The detection efficiency is defined by the ratio $\eta = \frac{N_\Omega}{N_{2\pi}}$, where $N_{2\pi}$ is the number of particles which are entering the detector surface from an isotropic point source and $N_\Omega$ are those particles which can be observed after etching, entering the detector in a solid angle $\Omega$ determined by the critical angle. Then the detection efficiency can be defined as:

$$\eta = \frac{N_\Omega}{N_{2\pi}} = \frac{\int_{0}^{\Omega} d\Omega}{\int_{0}^{2\pi} d\Omega}$$

but $d\Omega = \sin \theta \, d\theta \, d\phi$

$$\eta = \frac{1}{2\pi} \int_{\phi=0}^{\infty} 2\pi \int_{\theta=0}^{\pi/2 - \theta_c} \sin \theta \, d\theta \, d\phi$$

$$= 1 - \sin \theta_c$$

Substituting the value of $\theta_c = \arcsin \left( \frac{V_B}{V_T} \right)$

$$\eta = 1 - \frac{V_B}{V_T}$$

From the above equation it can be seen that numerous characteristics of these track etch detectors depend upon the track etch ratio $V_T/V_B$. The optimal detecting characteristic are obtained when $V_T \gg V_B$. In the ideal case when $V_T/V_B \rightarrow \infty$, $\theta_c \rightarrow 0$ and the detection efficiency $\eta$ becomes unity.
3.8. METHODS OF TRACK COUNTING

The following methods are used for counting the tracks in solid state nuclear track detectors.

3.8.1. By Optical Microscope

It is the most suitable method used for counting the tracks in SSNTD having the track density up to $10^5$ tracks/cm$^2$. After the chemical etching of a SSNTD, the tracks of particle can be easily counted by an optical microscope using ordinary magnification. In the case of normal or near normal incidence, the microscope can be focussed on the surface of the detector where the interaction of the conical tracks with the surface is observed as dark circular spots. By little defocussing, one can also see into the depth of the track. The tracks can be easily distinguished from the background scratches, etc. in the detector. Optical microscopy has been found to be highly useful for obtaining track densities and for studying some gross features of an etch-pit. The technique has limitations when a detailed structural analysis of an individual etched channel is desired.

3.8.2. By Naked Eye

Rare tracks of heavy and super-heavy nuclei in cosmic rays are usually detected by etched through tracks and can be visible
by the naked eye. This method was suggested by Fleischer et al. (1966). In this method an opaque coating of aluminium on one side of the plastic sheet having a thickness less than the expected length of the particle track is applied. On etching the plastic from uncoated side in a hydroxide solution, the hydroxide makes hole at the radiation damaged places and attacks the aluminium film. This attack is very rapid in short time, each track is surrounded by a relatively larger, easily visible circular area from which the aluminium has been removed. Using this method, they counted 100 tracks in 0.093 square meter of SSNTD.

Cross and Tommasino (1967) developed an alternative method of detecting holes in thin plastic sheets. This method may be used upto $10^3$ tracks/cm$^2$. Another method describe by Block and co-workers (1969) in which ammonia vapour penetrate the holes to make replica of the track pattern on a sensitized paper. This method could be applied to the hole as small as 0.7 μm in thin sheets of plastic.

3.8.3. **Gas Flow or Ionic Measurement Method**

Fleischer and Price (1963) proposed a method for track counting without the use of an optical microscope. In this method they used physical measurements such as gas flow or ionic permeability for the tracks counting through the irradiated material. Bean et al. (1965) developed such a method taking
mica as an irradiated specimen, which acted as a barrier between two halves of cell having a solution of hydrogen fluoride. Using this method, they got some reproducible curves in which conductivity was found to be a function of time for several samples of mica, whereas, Mory and Walker (1964) could not get reproducible curves while using irradiated plastics as barrier rather than mica.

3.9. AUTOMATIC METHODS OF COUNTING

The visual counting of etched tracks in solid state nuclear track detectors by an optical microscope is a difficult, time-consuming and expensive method. This problem is clearly found in, for example, absolute fission rate measurements, where the whole area of the exposed track detector has to be scanned (Azimi-Garakani and Williams, 1977). It was thought desirable to establish a rapid and simple method of track counting. Khan and Durrani (1972) set-up a electronic counting and projection system of etched tracks in SSNTD. The principle of the technique follows:

The detectors are exposed to fission fragments and then etched long enough to produce through or nearly through holes. The exposed detector then placed between an alpha source and a surface barrier detector. The etched fission tracks provide an easy path to the alpha particles which can not pass through unirradiated portion of the foil. The transmitted alpha
particles are registered by the surface barrier detector. The number of α-counts recorded by surface barrier detector is actually the number of fission tracks in the detector. The alpha particle used as a probe to measure the fission fragment track density. Since then a number of automatic counting systems of track in SSNTD have been developed, some of which are discussed below.

3.9.1. Spark Counting System

The spark counting technique was first devised by Cross and Tommasino (1970). This technique is very common in most of the laboratories because it is efficient, fast, reliable and does not require highly expensive and sophisticated equipment (Monin, 1980). It is specially useful for counting very low track densities. The technique works on the following principle.

A thin detector (~10-20 μm thick) in which the tracks are etched through is placed between a flat high voltage electrode and a thin aluminium electrode. The thin Al-electrode is an aluminized polyester film such as aluminized Myler. Through an RC network a high dc voltage is applied and causes a spark to occur through one hole in the film (Figure 3.7). The energy carried by the spark is sufficient to evaporate a larger area (<0.1 mm in diameter) in the thin Al-electrode, leaving the corresponding hole electrically isolated, which inhibits the
Figure 3.7. Basic set-up for the jumping spark technique.
occurrence of further spark in it. The spark, therefore, passes through another hole, jumping from one hole to another until it has passed through all the holes, and the discharge then stops. As a result, the thin Al-electrode, once removed, exhibits the distribution pattern of the tracks on the original detector and particularly allows their counting by eye, if not too numerous. But, more interesting, the current pulse of the sparks can be counted directly by scaler and in a matter of seconds provides the measurement of the track density. The design of spark counter and spark circuit have been given in Figure 3.8.

To get the reproducible results a very careful etching process should be performed and the residual thickness of the detector should be kept between two limits. The upper limit is the maximum thickness required to get etched-through holes from the particles. The lower limit is the minimum thickness below which the sheet can not withstand the applied voltage. Some other precautions regarding the spark counter are discussed by Monin (1980).

The total number of tracks that can be counted by this technique is limited primarily by the overlapping of the evaporated holes in the Al-electrode. Most of the time the maximum track densities reported are 3000 or 5000 cm\(^{-2}\) but can be as high as 15000 cm\(^{-2}\).
Recently a new automatic spark counting system is developed by Azimi-Garakani et al. (1981) which provides a convenient, cheap and fast method for absolute track counting measurements. The system is capable of obtaining reliable results for track densities up to $2 \times 10^4$ track cm$^{-2}$ for normal incidence irradiations. The counting efficiency of the system is about 97% compared with an optical microscope.

3.9.2. **Electrochemical Etching (ECE).**

The electrochemical etching technique enlarges etch-holes to a size of $\sim 100-300$ $\mu$m, which (for low track densities) makes their counting much easier. The technique was first suggested by Tommasino (1973) and then by other workers (Somogyi, 1977; Al-Najjar et al., 1978; Durrani, 1978; Al-Najjar et al., 1979). Figure 3.9 shows the apparatus which allows to etch the track electrochemically. The irradiated plastic detector is placed between two transparent cells containing the etching reagent, e.g. NaOH. A platinum electrode is placed in each half of the cell, and these are connected to a high frequency oscillating voltage supply (Durrani and Al-Najjar, 1980). Typically, peak voltages produce fields of $\sim 10-50$ kV cm$^{-1}$ across the plastic detector; frequencies of several kHz to tens of kHz are often used. High electric fields build-up in the vicinity of etched-tracks tips, leads to sparks. These yields Lichtenberg-type figures, which look
Figure 3.9. Schematic diagram of an electrochemical unit.
like sections through a carnation flower.

A good contact between the plastic detector and the two containers of the reagent, ensures electrical isolation of the two electrolytes. When filling electrolyte cells, efforts are made to exclude air from the film surface to avoid air bubbles.

The electrochemical etching technique is being extensively developed at present. The technique is widely applicable to neutron, proton and alpha particles measurements (Al-Najjar, 1979) with plastic track detectors.

3.9.3. **Image Analysis System**

Several image analysis systems are now available for the automatic scanning and counting of tracks in detectors. Some of those automated, computer driven devices are commercially available (Quantimet, Classimat, Leitz T.A.S.) and have been found suitable in SSNTD and nuclear emulsion (Fleischer et al., 1975; Azimi-Garakani and Williams, 1978). The principle involved is as follows:

The etched track detector is scanned by a closed-circuit TV camera to distinguish the features of interest from the general surroundings. For this purpose, a 'grey level' is so selected that only regions of the image where the blackness exceeds this pre-defined lower limit are considered. This enhances the contrast of the image. Shape discrimination can
be carried out with the help of extra modules which control different image parameters.

At present, a number of image analyser are available with microcomputers (e.g. Quantimet 900) which control not only the necessary steps in the analysis of the image, but also collect and analyse the data automatically and print out the results in any desired form (e.g. histogram; graph; etc.) (Durrani, 1982). Semi-automatic counting and size measurements of tracks can be carried out with the help of a video position analyser (VPA) interfaced with a micro-processor and a digitizing tablet. Other microprocessor based systems have been developed whose picture analysis is controlled by software (Heinrich et al., 1982).

3.10. ENVIRONMENTAL EFFECT

The solid state nuclear track detectors are amongst the least sensitive detectors to environmental effects. Most of the mineral insulators and glasses are relatively insensitive to several working conditions. Polymers are more sensitive but usually the environmental effect within normal variations can be ignored except under very extreme conditions. Factors which can cause significant effect on these detectors usually can be either controlled or be avoided.

Polycarbonate retains tracks up to a temperature at which it becomes viscous. Higher temperature can affect more sensitive
detectors such as cellulose nitrate. Exposures to gases and vapours such as $O_2$, $O_3$, $H_2O$ etc. during irradiation can increase the track etching rate of polymers (Boyelt et al., 1970). In a vacuum environment, the lack of these molecules decreases the etching rate $V_T$ and hence lowering the etching efficiency. Probably these molecules diffuse along the damaged region and attack the active species which make-up the track. The broken polymer chains would be prevented from reuniting and lessen the extent of damage produced.

Normally plastic track detectors are insensitive to beta and gamma radiations. But cross-linking and other effects of polymer degradation can also occur when the detectors are irradiated with heavy doses of gamma radiation. On an average 10 to 100 mrads are typically needed to produce any noticeable effects. Exposure to UV in presence of gases like $O_2$ or NO can affect the etching rate considerably. Table - 3.6 gives the summary of some important environmental effects.
### Table - 3.6

Summary of some Environmental Effects

<table>
<thead>
<tr>
<th>Effect</th>
<th>Polycarbonate</th>
<th>Cellulose nitrate</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Lexan</td>
<td>Makrofol</td>
</tr>
<tr>
<td>Thermal 1 hr Annealing</td>
<td>Total fading</td>
<td>185</td>
</tr>
<tr>
<td></td>
<td>50% fading</td>
<td>170</td>
</tr>
<tr>
<td></td>
<td>Start fading</td>
<td>40</td>
</tr>
<tr>
<td>Chemical</td>
<td>Add $N_2$, $V_T$ increases</td>
<td></td>
</tr>
<tr>
<td></td>
<td>Add $O_2$, $V_T$ increases</td>
<td></td>
</tr>
<tr>
<td>Irradiation</td>
<td>1.3 MeV, 200 Mrad $\gamma$, $V_T$ doubled</td>
<td></td>
</tr>
<tr>
<td></td>
<td>1.55 MeV, 25 Mrad electrons, $V_T$ increases</td>
<td></td>
</tr>
<tr>
<td>Photochemical</td>
<td>$O_2 + UV$, $NO + UV$ $V_T$ increases</td>
<td></td>
</tr>
<tr>
<td></td>
<td>$H_2O_2 + UV$, $N_2O + UV$ $V_T$ decreases</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
REFERENCES