CHAPTER - II

SOLID STATE NUCLEAR TRACK DETECTORS
2.1 INTRODUCTION

During the last two decades the Solid State Nuclear Track Detectors (SSNTDs) have found applications in almost all branches of science and technology. SSNTDs are the most widely used detectors for low as well as high energy ionography. They include the dielectric solid materials mainly natural, volcanic and man made glasses (phosphate, soda lime, tektite, silica, flint, granite, obsidian etc.), minerals (allanite, apatite, aragonite, garnet, sphene, zircon, quartz, epidote, feldspar, diopside, mica etc.) and plastics (cellulose acetate, cellulose nitrate, cellulose propionate, cellulose triacetate, polyster, polyamide, polycarbonate, polyethylene, polystyrene, polyethylene terephthalate and Columbia Resin (CR-39) Plastics etc.). Basically SSNTDs are classified as inorganic crystals, inorganic glasses and high polymers (plastics). Plastics, the most sensitive group have wide applications in the study of heavy ions, cosmic rays, neutron dosimetry and radon dosimetry etc.

The principle of detection of charged particles by the SSNTDs is based on the fact that the passage of heavily ionizing nuclear particles through most dielectric solids produce narrow intense damage trails on an atomic scale (~ 30-100 Å) along the trajectory, as a result of the excitation and ionization of atoms. In crystals, the latent trails consist of atomic displacements whereas in
plastics or polycarbonate the damage is due to broken molecular chains which produce free radicals etc. The damaged narrow region of the solid having different chemical and physical properties than the undamaged bulk material of solid is called the 'Latent Track'. The latent track can be seen directly only with Transmission Electron Microscope. However, it can be seen under optical microscope after enlarging upto few microns through chemical etching. As the damaged region of the solid dissolves faster in an appropriate etchant as compared to undamaged material, these latent tracks may produce conical etch pits or in certain cases narrow conical channels called 'Tracks'. When these conical etch pits are enlarged to sizes comparable to the wavelength of visible light, they can be observed under an optical microscope at ordinary magnifications.

Now-a-days the SSNTDs have grown to such an extent that there is hardly a branch of science and technology where these do not have an application. As compared to other detectors such as bubble, cloud, spark chambers and even nuclear emulsions etc. these are extremely simple to construct. Basically, almost all of the materials used in etched track ionography are fabricated as electrical insulators and for use in the manufacture of a wide range of everyday objects (Durrani and Bull, 1987).

2.2 HISTORICAL FEATURES

In 1959 E.C.H. Silk and R.S. Barnes first observed the damage
trails produced by fission fragments at the British Atomic Energy Research Establishment at Harwell (England). Their observations gave a chain of ideas about the discovery of particle tracks and have helped to create an aura of excitement and fascination for particle tracks in solids.

Basically, the era of nuclear tracks began in 1958 when D.A. Young working at the Atomic Energy Research Establishment at Harwell published a remarkable note in Nature (Young, 1958) on the presence of etch pits in LiF crystals. According to which the crystals were chemically etched following their irradiation with thermal neutrons along with a uranium foil kept in close contact with them, to reveal fission fragment tracks. He also briefly described possible mechanism of track formation whose principle is identical to what some models later proposed for track formation mechanism in inorganic detectors. Further analysis of the crystals showed a complete correspondence between the number of etch pits and the estimated number of fission fragments which would have recoiled into the crystals from the uranium foil. One year later Silk and Barnes working in the same institute reported the direct observation of those nuclear tracks in mica using Transmission Electron Microscope (TEM).

The extensive development of this technique of observing
nuclear tracks in dielectric solid crystals and glasses was done by a team of scientists (Fleischer, R.L., Price, P.B. and Walker, R.M.) at the General Electric Research Laboratory at New York. They repeated the observations of some earlier workers. Successively, they also observed that the damaged trails in mica can be etched preferentially in an appropriate etchant and can be observed under an optical microscope (Price and Walker, 1962a; 1962b). Fleischer and Price (1963a,b) also pointed out that the spontaneous fission of the trace amounts of uranium present as impurities in rocks would leave such type of latent trails in crystals of the rock. They also indicated that the nuclear tracks could be etched and observed in glass, minerals as well as in plastics. Fleischer, Price and Walker thus established this new field of 'Trackology' and put the ideas on the possibility of applications of SSNTDs in science and technology. Due to their outstanding properties like simplicity, flexibility, durability and their specific nature of response to various radiations, they found rapid applications in diverse fields of science and technology. After the first review paper of Fleischer et al., (1965a) the scientists started working on SSNTDs and their applications in different laboratories of the world. Thus, the growth of fundamental knowledge and technological applications of 'trackology' based on ionographic registration in SSNTDs established.
2.3 IMPORTANCE OF SSNTDs AND THEIR PLACE IN PARTICLE DETECTION

The track formation in SSNTDs is related to the ions which produce damage in a solid, exceeding certain threshold unit so that damage concerned is permanent along the trail of the ion. This property of track etch detectors is one of the main advantages in particle identification. The threshold for the detection of a particle is quite favourable property when one has to detect a heavy particle and avoid an intense background of ionizing radiations. The intensity of ionization damage produced by an ionizing particle is directly proportional to the square of its charge and approximately inversely proportional to the square of its velocity. This property of charged particles for detector material has been used successfully by various workers in the study of cosmic ray particles through nuclear track registration. Sensitivities to nuclear particles for different materials have been found different for different ions theoretically as well as experimentally. Plastics (organic polymers) are found to be the most sensitive. Some well known and commonly used plastics and their relative sensitivities are cited in Table 2.1.

The threshold characteristic is an important tool in many applications in physical sciences and contributed significantly to the establishment of the solid state nuclear track detectors. SSNTDs have many useful features over other nuclear charged particle
Table 2.1
Relative sensitivities of some commonly used plastics

<table>
<thead>
<tr>
<th>Plastics</th>
<th>Chemical Composition</th>
<th>Least observable ion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Allyl diglycol carbonate (CR-39)</td>
<td>$\text{C}<em>{12}\text{H}</em>{18}\text{O}_7$</td>
<td>10 MeV $^1\text{H}$</td>
</tr>
<tr>
<td>Amber</td>
<td>$\text{C}_2\text{H}_3\text{O}_2$</td>
<td>Fission fragments</td>
</tr>
<tr>
<td>Bisphenol-A polycarbonate (Lexan, Makrofol)</td>
<td>$\text{C}<em>{16}\text{H}</em>{14}\text{O}_3$</td>
<td>0.3 MeV $^4\text{He}$</td>
</tr>
<tr>
<td>Cellulose nitrate (Diacel)</td>
<td>$\text{C}_{6}\text{H}_8\text{O}_2\text{N}_2$</td>
<td>0.55 MeV $^1\text{H}$</td>
</tr>
<tr>
<td>Cellulose nitrate (LR-115)</td>
<td>$\text{C}_{6}\text{H}_8\text{O}_2\text{N}_2$</td>
<td>0.1 MeV $^1\text{H}$</td>
</tr>
<tr>
<td>Polyethylene</td>
<td>$\text{CH}_2$</td>
<td>Fission fragments</td>
</tr>
<tr>
<td>Polymide</td>
<td>$\text{C}_{11}\text{H}_4\text{O}_4\text{N}_2$</td>
<td>36 MeV $^{16}\text{O}$</td>
</tr>
<tr>
<td>Polyoxymethylene (Delrin)</td>
<td>$\text{CH}_2\text{O}$</td>
<td>28 MeV $^{11}\text{B}$</td>
</tr>
<tr>
<td>Polypropylene</td>
<td>$\text{CH}_2$</td>
<td>1 MeV $^4\text{He}$</td>
</tr>
</tbody>
</table>
detectors (ionization chamber, scintillation counter, cloud chamber, Geiger Muller counter, proportional counter, nuclear emulsion, bubble chamber, spark counter and semiconductor detectors etc.).

Some important features of solid state nuclear track detectors are enumerated as follows:

i) These detectors are simple in construction and can be obtained in very small as well as in very large sizes.

ii) These are cheap and can be conveniently used.

iii) These are insensitive to light, X-rays, \( \gamma \)-rays and \( \beta \)-rays etc.

iv) These detectors record tracks permanently 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 (Fleischer et al., 1965b).

v) These solids being robust do not require very careful handling.

vi) These are threshold type detectors and the best utilization of this property is for the detection of heavy ions such as fission fragments which can be recorded and distinguished from a very high background of light charged particles like \( \text{^{4}He} \), \( \text{^{2}H} \), \( \text{^{1}H} \), \( \beta \)-particles, X-rays, \( \gamma \)-rays and even neutrons.

vii) A very high efficiency and sensitivity can be obtained if the detectors are placed in direct contact with the fission
viii) These detectors also possess the charge and energy discrimination properties. It has recently been reported that the resolution for high z particles obtained with plastic detectors is better than nuclear emulsions.

ix) These detectors also have a considerable amount of geometric flexibility and are, therefore, particularly useful in angular distribution measurements.

x) To study nuclear interactions in these detectors high spatial and time resolutions may be achieved using electron microscope.

xi) The revelation of tracks in these detectors through preferential chemical etching process is very quick and simple as compared to nuclear emulsions.

xii) Out of SSNTDs some solids themselves can be used as targets as well as detectors viz. plastics, while measuring neutron flux recoil nuclei and glasses and mica etc. in neutron fluence measurements using fission tracks. Fission tracks arise due to the impurities present such as $^{235}$U.

Despite these advantages, these SSNTDs suffer from some disadvantages with them given below.

i) Only a part of the total track length of the particle trajectory can be seen leaving the part that has been etched
ii) Although charge and mass resolution of charged particles has been possible with these detectors, an accurate energy resolution has still not been possible.

iii) One has to perform a blind experiment as during the experiment, the monitoring of the experimental observations is not possible.

After comparison of the properties of SSNTDs with other detectors, it can be easily observed that SSNTDs have many advantages over other detectors in general. Light charged particles such as $^1\text{H}$, $^2\text{H}$, $^3\text{He}$, and $^4\text{He}$ were detected by the use of SSNTDs and attempts were also made to make some reasonable estimates of their energies. But later they have found wide applications in heavy ion and cosmic ray studies.

2.4 APPLICATIONS OF SSNTDs

The track registration technique of solid state nuclear track detectors has found wide and diverse applications in almost all fields of science and technology and there is a great fascination towards them. A brief discussion of the important applications of SSNTDs is given below.

2.4.1 Nuclear physics

Particle identification characterised by their charge $z$ and
mass \( m \) is a major area of applications of SSNTDs in nuclear physics. The tracks formed in the detector medium depends on the properties of the incident particle namely its energy \( E \), velocity \( v \) and rate of energy loss \( (dE/dX) \) in the medium. The total energy of the particle (function of mass and velocity) and the nature of the particle can be precisely determined if one can precisely scale the rate of energy loss which is a function of incident particle charge and velocity. They are ideal for fission studies because SSNTD technique can provide free detection of heavy charged particles (such as fission fragments) in the presence of high dose of light rays, \( X \& \gamma \)-rays and also light charged particles (such as \( \beta \)-particles) and neutrons etc.

Plastic track detectors are also used for charge and mass resolution. Price et al (1970) obtained a charge resolution \( \Delta z \) of \( \sim 0.3 \) charge unit and mass resolution of \( \sim 2 \) amu using Lexan. In cosmic rays studies a mass resolution of 0.7 amu for Fe isotopes was reported (Siegman et al., 1976) using UV sensitized Lexan. A \( \Delta z \) of \(<0.1 \) charge unit and \( \sim 0.3 \) amu mass resolution in CR-39 (Cartwright et al., 1978) are also reported. SSNTDs have been successfully used for the determination of half life of spontaneous fissioning nuclei. It is also possible to obtain the half life by comparing with the number of tracks produced by a natural radioactive substance with half life comparable with that of unknown radionuclide. To determine
compound nucleus life time by blocking effect is another remarkable application of track detectors in nuclear physics. Gibson and Nielson (1970) measured the life time of $1.4 \times 10^{-6}$ sec for the decay of the $^{238}$Np compound nucleus in the first successful experiment. This was accomplished by bombarding $\text{UO}_2$ single crystal with protons of two different energies and by comparing the blocking patterns of fission fragments emitted from the crystal imaged in a Makrofol detector. For the measurement of half life of alpha emitting compound nuclei, alpha sensitive track detectors can be used.

Microanalysis of fissionable trace elements is another potential application of SSNTDs. Micromapping upto sub-ppb level is possible with this technique. The detailed description of the technique is given in the last chapter of this thesis. Some other important applications of SSNTDs in nuclear physics are:

i) The determination of fission barrier and saddle point mass of nucleus (Burnett et al., 1964).

ii) The search and determination of half lives of Super Heavy Elements produced by galactic nucleosynthesis during the formation of solar system. Bhandari et al. (1971) has claimed to have obtained fossil track evidence for Super Heavy Elements.

iii) Ternary fission was first observed by Fleischer et al.
A number of track detectors are used in the investigations on the ternary photofission (Medveczky and Somogyi, 1970). Methasiri and Jahansson (1971) have detected high energy photofission of heavy and medium heavy elements.

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

v) Synthesis of element \( z = 104 \) was demonstrated by Flerov et al. (1965).

vi) For the study of fragments emitted in radioactive heavy ion reactions.

vii) Track detectors (eg. Lexan) are used to determine angular distributions, energy spectra and charge distribution of low energy fragments and also in relativistic heavy ion reactions.

viii) Angular anisotropy and nuclear pair correlation effects in nuclear fission were observed by Smirenkin et al., (1968).

ix) In 1973, Chung had determined the cross-section for the electrofission of \( ^{200}_{\text{Hg}} \).

x) Some rare multiprong events were observed (Brandt et al., 1980; Khan et al., 1980).
2.4.2 Heavy ion research and related studies

SSNTDs have many important applications in research works related to charged particles. Because these detectors are insensitive to X and γ-rays and also have threshold property for detection, they are used widely in heavy ion research such as composition of cosmic rays, heavy ion nuclear reactions, radiation dose due to heavy particles, explorations of super heavy elements and magnetic monopole etc. (Fleischer et al., 1975). Dial et al. (1955) predicted that the pattern of energy loss along the trajectory of fission fragments was entirely different from the pattern of energy loss followed by energetic heavy ions (energies exceeding about 5 MeV/n) such as Fe, Pb, Xe and U ions. The values of the rate of energy loss and the ranges of such ions are not very accurately known even though the heavy ion accelerators have made available various heavy ions having energies exceeding 5 MeV/n or so. However, recently some calculations have been made based upon certain assumptions which incorporate values of the interaction parameters used for the interactions involving light ions viz protons and alpha particles (Chohan et al., 1988). They showed that the calculated values have poor agreement with those observed experimentally. Therefore, it has been realized that experimental determination of the ranges of energetic heavy ions in various media of interest is a matter of great importance. For example, it is possible to measure the rate of energy loss, total energy and
velocity of a particle with sufficient accuracy. Then, since $\frac{dE}{dX}$ in the given medium is a function only of the particle charge and velocity and the energy is a function of its mass and velocity, the nature of the particle may be precisely determined.

The determination of the charge spectrum of the heavy nuclear content of the primary cosmic rays is one of the earliest fields of application of plastic detectors. The necessity to lift the detector above the bulk of the atmosphere which so effectively shields the earth from the cosmic rays, places severe constraints on the complexity of the detecting systems that can be used. Thus the dielectric detectors are particularly useful in these type of studies. The stacks of nuclear emulsions were used in the earliest studies of heavy primary cosmic rays (Fowler et al., 1967; 1970). However, these have now been largely superseded by plastic track detectors. In the studies of ultra-heavy cosmic rays, where very large detector areas are to be used, these detectors have demonstrated their utility.

2.4.3 Radiation dosimetry

The solid state nuclear track detectors have been widely used for the measurement of the amount of radiation which basically determines the dose of ionizing radiation absorbed by a body or some parameters in correspondence to the absorbed dose. For an ideal dosimetry system, especially a personal dosimeter, it should be
robust, simple, cheap and easily available. Having all these properties the SSNTDs are finding increasing application in radiation dosimetry. Radiations usually encountered in dosimetry are α-particles and heavy ions which have sufficiently high linear energy transfer (LET, the energy lost by particle per unit path length) to be detected by dielectric track detectors directly. Neutrons and energetic pions can also be recorded by generating high LET secondary radiations in passing through matter. Neutrons, being electrically neutral, are detected through their nuclear reaction products (Walker et al., 1963). Detection of thermal neutrons usually utilizes $^{235}$U which undergoes fission by thermal neutrons. With a thick foil of almost pure $^{235}$U and fission fragment detector like Lexan policarbonate, thermal neutrons dose upto 0.1 mSv (equivalent to a thermal neutron fluence of $10^4$ cm$^{-2}$) can be practically measured (Durrani and Bull, 1987). At higher energies, neutrons transfer significant amount of energy and, therefore, their relative biological effectiveness (RBE, expressed as quality factor) are also high. By the use of a simple dielectric track detector, it is not possible to obtain information about the neutron energy but a measure of total dose equivalent can be obtained. To determinate and record a wide range of neutron energy, a two detector assembly incorporated with natural and enriched uranium was desired (Tatsuta and Bingo, 1970). In the devices using fission foils, the major disadvantage is their inherent radioactivity which can deliver a
significant dose to the persons who wear it. The unique features of solid state nuclear track detectors used for neutron dosimetry are

i) Insensitivity to background effects due to β, X, and γ-radiations.

ii) These detectors record tracks permanently and without environmental effects.

iii) These have no requirement of immediate retrieval and processing.

iv) Non-requirement of an electronic counting equipment for evaluation.

v) Wide range of doses can be studied.

vi) Non-requirement of dark room condition for the development and fixing of the latent image as in the case of nuclear emulsion.

Alpha dosimetry using SSNTDs is simple as compared to neutron dosimetry because alpha particles can form etchable tracks in the detector directly. However, the range of alpha particle energies over which this will leave tracks in detector is limited by threshold rate of energy loss above which tracks are formed. Radon gas and air-borne radon progeny may be the naturally occurring hazardous alpha particle sources. SSNTDs are widely used throughout the world for various measurements related to radon and its progeny dosimetry.
2.4.4 Geochronology and archaeology

As fission fragments from the spontaneous break-up of $^{238}\text{U}$ are capable of registering chemically etchable tracks in most of the rock forming minerals and uranium is a fairly ubiquitous trace element at the $\sim 1 \mu\text{g/g}$ to $10^{-3} \mu\text{g/g}$ (1 ppm to 1 ppb) level in geological materials, it opens up the possibility of a widely adopted technique of dating of rocks (Price and Walker, 1962c). Naturally occurring crystals, glasses and rocks are found to have recorded tracks in them. Being insensitive to light charged particles and ionizing radiations produced in $\alpha$ or $\beta$ decay, these internal tracks should be entirely due to either fission or recoil nuclei (Fleischer et al., 1975). The natural tracks originated from the spontaneous fission of natural heavy radionuclides ($^{235}\text{U}, ^{232}\text{Th}, \text{Bi, Pb, etc}$) and fission of heavy elements induced by cosmic rays and spallation caused by cosmic ray secondaries etc. can be identified.

The density of the 'natural' damaged track trail, is given by

$$\rho = k \cdot c \cdot \text{Age}$$

where $c$ is the uranium content of the sample; Age, the age of the sample and $k$, a constant which is the function of the geometry of the surface analysed and of the range of the fission fragments in the material. Therefore, the 'age' of the sample can be determined.
by comparing the induced track density. Of various radiometric
dating, the fission dating is conceptually simple. $^{238}\text{U}$, the major
isotope in natural uranium, decays by spontaneous fission at a rate
of $\sim 10^{-16}$ per year. Therefore, a mere estimation of the fraction of
the uranium atoms that have fissioned within the sample can give an
estimation of its age. The technique of fission track dating has
been extended to investigate ocean bottom spread, the continental
drift and dating of geological, archaeological and cosmological
samples (Fleischer, 1975). In place of $^{238}\text{U}$ content, it is more
convenient to measure the $^{238}\text{U}$ content of the sample and considering
the abundance ratio of $^{235}\text{U}$ to $^{238}\text{U}$ constant (as is almost the
case), the determination of $^{238}\text{U}$ content by fast neutron fission is
avoided as these neutrons would also produce fission in the $^{232}\text{Th}$
content of the sample. Extremely interesting informations regarding
the man period and the history of seafaring near the Greek Main bond
have been obtained through the track studies of natural 'Roman
Glasses' and the 'Franchtic cave' obsidians (Durrani et al., 1971).

2.4.5 Biological applications

SSNTDs are extensively used in biological applications
(Durrani, 1982) e.g. inhalation of $\alpha$-active aerosols, filtration of
malignant cells by microporous filters and the measurement of
environmental $\alpha$-emitters (used in radiation dosimetry). Recently,
main interest has been taken in the radiological consequences of the
inhalation of α-active particles and their deposition in lung through tobacco smoke or in the atmosphere of uranium mines. Track detectors have been used in the radiobiology. Track registration technique has been employed for mapping the locations of plutonium concentrations in living matter.

Nuclear Track Filters (NTFs) have been used in various fields including microbiology, virology, medical science, aerosol science and food processing (Fleischer et al., 1975). NTFs made out of many plastic detectors using normally incident heavy energetic ions are reported in the literature (Fischer and Sphor, 1983; Chakarvati et al., 1986). For this purpose a thin detector material is irradiated to a collimated beam of ions that produce tracks across the entire thickness of the sheet and one then etches to produce holes that perforate the membrane or filter. The hole diameters can be controlled by specifying the etching conditions and time, and the density of holes is controlled by incident ion dose. Holes with diameters ranging from 50 Å onwards can be produced in mica and various plastic materials. Etched track filters have proved useful in environmental studies of particulates in the atmosphere and oceans.

2.5 TRACK FORMATION MECHANISMS

2.5.1 Theory of track formation

The track formation or production means the process by which
the charged particle alters the solid along its path to produce damaged trail. Ionizing particles passing through dielectric materials create intense trails of damage on atomic scale. The damage produced by irradiation of solids depend upon the intrinsic properties of incident particles like mass, charge, velocity and the composition of the material of the detector itself. Particle tracks are formed in many insulating materials and some semiconductors. The charged particle latent tracks in solids are narrow (\(<50\, \text{Å}^0\) radius), stable and chemically reactive centres of strain that are composed mostly of displaced atoms rather than electronic defects (Fleischer et al. 1975). The heavy ion deposits energy while slowing down in a solid and it is very important in understanding the track formation mechanism. The track registration property of the detector depends upon the sensitivity of material. Solids with resistivity more than \(2\times10^3\, \text{ohm.cm}\) generally store tracks.

Different type of materials have different critical rates of energy loss for track formation (Fleischer et al. 1964; 1965b). In plastics, ionizing radiations directly produce ionized and excited molecules and electrons. Some excited molecules may de-excite through the emission of radiation or through non-radiative transitions. Excitation energy can also be transferred from one molecule to another. Electrons are trapped at various sites or can combine with molecules to form negative ions or recombine with
positive ions yielding excited molecules. Both ions and excited molecules may acquire considerable vibrational energy and undergo bond rupture to form a complex array of stable molecules, free radicals and radical ions.

In case of inorganic crystals, the effect of radiation upon the material is to produce ionization and excitation of atoms or molecules. Electrons are raised across the forbidden energy band. Some of these may return to the valence band via luminescence centres with the emission of radiation, while others after diffusing through the crystal will either be trapped at the sites of various imperfections or will return via non-radiative transitions to positive ions. Low energy heavy ions will produce numerous atomic displacements directly through elastic collisions. Electron irradiation can also cause direct atomic displacements. An electron of rest mass $m_0$ and having kinetic energy $E$ can transfer a maximum energy $E_{\text{max}}$ to an atom of mass $M$ given by

$$E_{\text{max}} = \frac{2(E + m_0 c^2)E}{M c^2}$$  \hspace{1cm} (2.5.1)

For example, an electron of energy 1 MeV can transfer energy up to ~ 100 eV to a silicon atom, whereas only ~ 25–30 eV is required to produce displaced atoms. The number of atomic displacements produced by electrons can exceed the levels as expected from direct
collisions between electrons and atomic nuclei (Varley, 1954). When a fast moving atom of atomic number \( z \) traverses the solid its orbital electrons interact with the electrons of the atom of which the solid is made off. The result of these interactions is that the moving atom would rapidly become an ion by being stripped of all or some of its orbital electrons. Therefore, the ion acquires a net positive charge \( z^* \) which can be empirically expressed (Heckmann et al., 1960) as

\[
z^* = z[1 - \exp(-130\beta/z^{2/3})]
\]

Where \( \beta (=v/c) \) is the speed of ion relative to the velocity of light. At high velocities where \( z^* = z \), the dominant interaction is due to the coulomb force between the ion and the electrons attached to the atoms within the solid. As a result of this interaction two effects occur:

i) To excite electrons to higher energy levels (excitation)

ii) To loose electrons from their atoms which can be stripped out of the atoms (ionization).

The ejected out electrons in atoms are called delta rays and can produce further excitation and ionization if they carry enough energy. Generally in inorganic solids primary ionization is dominant source of the track damage. The primary ionization occurs close to the path of the ion, while the secondary ionization and excitation
is spread over large radial distances from the core of the track, hence the effects of delta rays are unimportant. When the ion slows down in passing through solid, it eventually regains orbital electrons one by one as its velocity becomes comparable with the orbital velocity of less and less tightly bound electrons. At lower velocities (less than 50 KeV/amu), the atomic collisions become more dominant mode of energy loss. However, in organic polymers primary ionization as well as secondary ionization (delta rays) contribute to the formation of tracks.

2.5.2 Track formation mechanism models

To account for the track production and other characteristics of the track formation, different models have been proposed. Out of those some successful models are:

i) Thermal spike model

ii) Displacement spike model

iii) Ion explosion spike model

iv) Restricted energy loss model

i) Thermal spike model

According to this model charged particle first gives energy to the electrons of the solid then it is transferred to the atom by electron phonon collisions, causing thermal motion. Due to this the incident energetic particle dislocates the crystal lattice and produces strong heating in the part of the solid that is traversed
by the particle. This region is, therefore, raised to a high temperature and then cools rapidly via heat conduction to the surrounding materials. If the temperature is sufficiently high, it can cause permanent alterations thus producing tracks (Chadderton et al., 1963; 1966). This model failed to relate the sensitivities of different materials in any regular manner with a known melting, softening or transformation temperature of detectors (Fleischer and Price, 1964) and also could not distinguish satisfactorily among the materials in which tracks could be formed and those in which the tracks could not be formed.

ii) Displacement spike model

According to this model particle alters the solid along the path by displacing atoms in the particle nuclei collisions. But this model could not explain why the tracks were not formed in metals and hence it was rejected. According to it tracks should be more prevalent near the end of the range of energetic particles. It is due to the fact that the loss of energy by particle-nuclei collisions becomes more efficient as the velocity of particle decreases.

iii) Ion explosion spike model

Fleischer et al. (1965c) proposed a semi-quantitative model for track formation known as the 'Ion Explosion Spike Model'. This model is based on the postulate that a heavily ionizing particle leaves a
narrow region of high density positive charges and thus energy is first lost to the electrons of the atoms along its trajectory. As the time for electron positive ion recombination is longer compared with the lattice vibration time ($\sim 10^{-13}$ second), mutual repulsion can drive these ions into interstitial positions. Subsequently, the neutralization of the positive ions and relaxation of the surrounding lattice into the disrupted region take place and lattice strains are set up around the track core. The track formation in case of inorganic solid is explained as shown in figure 2.1. The incoming ion first knocks out electrons from the atom in its way, thus creating an unstable array of adjacent positive ions. Then the positive ions repel and thrust one another away from their normal sites into the interstitial positions in the crystal lattice, thus creating vacant lattice sites due to their coulomb repulsive forces.

In case of organic polymers or plastics the atoms are arranged in a chain like structure. The excited atoms produced by the incident charged particle lead to the breaking up of long molecular chains of the polymers. These broken molecular chains rarely reunite at the same place, rather they produce broken bonds and free radicals etc. Thus the paths of broken molecular chains are chemically more reactive (Figure 2.2).

According to the ion explosion spike model the quantitative
Figure 2.1: (a) The incident heavy ion produces primary ionization along its trajectory. Thus leaving an unstable array of +ve ions (b) the +ve ions so created repel and knock each other from their normal sites and move into the interstitial space in the crystal lattice causing electrostatically stressed region (c) the stressed region relaxes elastically and propagates the strain in side-ways.
Figure 2.2: Track formation in organic polymers (Plastics).
criteria for the requirement of track formation are as follows:

(i) For track to be formed the coulomb repulsive forces within the ionized region must be greater than the lattice binding forces i.e. the 'electrostatic stress' must be greater than the 'mechanical strength' of the material. If two atoms have received an average ionization of n unit charge e and are separated by a distance 'a₀', the force between them and the electrostatic stress are

\[
\frac{n^2 e^2}{4 \pi \varepsilon \varepsilon_0 a_0^2} \quad \text{and} \quad \frac{n^2 e^2}{4 \pi \varepsilon \varepsilon_0 a_0^4}
\]

respectively

where \( \varepsilon \) is the dielectric constant for the material, \( \varepsilon_0 \) is the permittivity of free space.

The total mechanical strength in terms of macroscopically measurable quantities can be obtained by equating it with mechanical tensile strength. According to Fleischer et al. (1975) the bonding force of the lattice (mechanical strength of the material) may be taken as \( Y/10 \) (= 0.1 Y) as a working approximation where \( Y \) is Young's modulus of the material. Thus the criterion for track formation may be expressed as

\[
\frac{n^2 e^2}{4 \pi \varepsilon \varepsilon_0 a_0^4} > 0.1 Y
\]
where $s$ is called the stress ratio (Fleischer et al., 1965c) of the material and is a measure of relative sensitivity of various track recording materials. This stress ratio shows a fairly good correlation with the sensitivity of track recording materials. The relation (2.5.3) indicates that the materials having smaller value of $s$ should have smaller values of mechanical strength $Y$, dielectric constant $\varepsilon$ and lattice spacing $a_0$ for being sensitive. For polymers the ratio is $s \approx 0.01$ whereas for inorganic crystals it is $\approx 1$. Therefore, plastics are more sensitive than inorganic crystals.

(ii) Secondly it shows that the track can be formed only if the electrons can not drain into the region from which the positive ions have been displaced by repulsion in less than one vibration time i.e. $10^{-13}$ second. If the positive ion core is to survive long enough ($\geq 10^{-13}$ second) for a track to form, the free electron density must be low. If the density of free electrons is $n_f$, then

$$n_f < \frac{e_n}{\pi a_0 \mu_e k T t}$$

where $\mu_e$ is the electron mobility, $T$ is the absolute temperature, $k$ is the Boltzmann's constant and $t$ is the diffusion time for
electrons i.e. lattice vibration time. This condition is satisfied by insulators and poor semiconductors but not by metals and explains that tracks will be formed in the insulators and semiconductors but not in metals.

(iii) The third condition for latent track formation is that the hole mobility should not be too high in order to avoid recombination of ionised atoms and electrons to get neutralised. The tracks will not be formed in those materials in which hole mobility is greater than $10 \text{ cm}^2/\text{V sec}$. In semiconductors and metals the hole mobility is too high at room temperature. Therefore, only insulators satisfy the condition for the track formation.

(iv) The fourth condition is that there must be at least one ionization event per atomic plane i.e. the damaged region by the incoming particle must be continuous to the atomic extent along the trajectory of charged particle for the most successful track revelation. Thus, the ion explosion spike model predicts the sensitivities of various track recording materials and accounts for the inability of metals and good semiconductors for revelation of tracks.

2.6 CRITERIA FOR TRACK REGISTRATION

Track formation validity of various models may be judged by critical appraisal of parameters such as charge, mass, and energy of such incident particles those are able to form etchable tracks.
Track formation can also be related to the total energy loss rate, primary ionization, restricted energy loss etc. of the ionizing particle. According to track formation criteria, considering these parameters, it was stated that tracks are formed in a medium when and only when, the chosen parameter exceeds some critical value, whatever be the bombarding particle. For example, the alpha particle can produce an etchable track in cellulose nitrate, cellulose acetate and polycarbonate plastics but not in polythene plastics like melinex, hostaphan, myler and terphan while the fission fragments are able to record tracks in these plastics. The relevance for the formation of track criterion is in the identification of ionising particle.

2.6.1 Total energy loss rate criterion

Fleischer et al. in 1967 proposed the dependence of total amount of energy deposited per unit path length on the track formation. It was suggested that in solids, only those particle will register the tracks which have a total rate of energy loss more than a critical value \( \frac{dE}{dX} \) needed by that solid. This quantity was considered to be the characteristic of the solid material.

Fleischer et al. (1967) calculated the value of \( \frac{dE}{dX} \) and plotted as a function of energy per nucleon for various heavy ions in the detectors viz. muscovite mica, lexan polycarbonate and cellulose nitrate. The data were consistent with the hypothesis that
for each solid there exists a critical rate of energy loss (dE/dX)_{crit.} such that particles losing energy more rapidly than this critical value produce continuous tracks with unit efficiency. The particles, depositing appreciably less energy per unit length can not produce tracks.

The main shortcoming of this criterion is that it completely neglects the primary ionization for the track formation.

2.6.2 Primary ionization criterion

Fleischer et al. (1967) suggest that the results on track registration measurements are inconsistent with total energy loss rate dE/dX criterion. Using the total energy loss rate criterion, they predicted the minimum detectable charge of 70 for relativistic ions in lexan. But experimentally it was found that relativistic ions of charge 57 ± 2 can produce etchable tracks in it. It is also known that the tracks are registered in all three solids (muscovite mica, lexan polycarbonate and cellulose nitrate) at energy loss rates below the predicted threshold and the relativistic iron nuclei should not form tracks in cellulose nitrate. All these descriptions led to the formation of a new criterion for track registration.

Ion explosion spike 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

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

Where $z^*$ 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 atoms and $\alpha$ is the constant that depends on the material.

Solid can record an etchable track only if the linear ion density produced by the primary particle along the trajectory is greater than a critical value for that material. This model also explains that a solid would register a track if the rate of primary ionization ($dJ/dX$) is greater than a critical rate of primary ionization i.e. $(dJ/dX)_{\text{crit}}$, which is a characteristic of the material. It was concluded (Fleischer et al., 1967) that for each solid it is possible to select a critical value of the primary specific ionization above which tracks are formed and below which there is no preferential etching for track formation in that solid.

The secondary ionization produced by delta rays occurs almost along the side of the trajectory 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 and Nix (1969), according to which the low energy delta rays play a vital role in track formation mechanism in polymers. According to the ion explosion spike model it is expected that only the number and not the energy of the removed electron is of great importance as long as the electrons receive enough energy to separate them out of the track region.

The main objectives to critical primary ionization criterion, are:

(i) The absolute values of \((dJ/dX)_{\text{crit}}\) are not known because the values of dimensionless constants of stopping materials as used by Fleischer et al. (1967) are strictly true for hydrogen.

(ii) The effect of secondary ionization of delta rays has been neglected.

(iii) 2 eV energy utilized as the ionization energy of the outer electron of the atoms in plastics appears to be inconsistent with the ion explosion spike mechanism which requires ejection of electron from the track region. Generally, 2 eV energy is not sufficient to ionize the atom but it may be enough to break a chemical bond. Bovey (1958) suggested that for ionization, a higher value of 9 - 15 eV is required. Therefore, this criterion is not found to be suitable for track registration.
2.6.3 Critical restricted energy loss rate criterion

Benton and Nix in 1969 proposed a new track formation criterion for plastic track detectors known as 'Restricted Energy Loss' criterion. In view of the objections to primary ionization criterion they incorporated the secondary ionization and excitations produced by the low energy recoil electrons (low energy delta rays). According to this criterion the total energy deposited per unit track volume by the incident charged particles determines the chemical reactivity of the latent trail region. It was predicted that a relativistic heavy ion having high energy produced delta rays along the particle trajectory. These delta rays having several MeV of energy tend to be scattered and deposit their energy at a considerable distance (on atomic scale) from the path of heavy ion. In case of plastics (generally 100 μm thick), these energetic delta rays and most of their accompanying energy are lost from the detector and, therefore, it is not considered in track formation. Thus, the total rate of energy loss (dE/dX) of a charged particle can not be used as criterion for track registration. This leads to the invalidity of the criterion of total energy loss rate (dE/dX) for track formation.

The restricted energy loss rate criterion in addition to the primary ionization, considers that the secondary electrons having the energy \( w \) less than a predetermined value \( w_o \), contribute to the
track formation. If the energy density is greater than the minimum value depending upon the nature of the detector, then only a particle can register an etchable track in the detector.

For calculating the value of \( (\frac{dE}{dX}) \) \( _{w<w_o} \) (Benton and Nix, 1969) used the relation,

\[
\frac{dE}{dX} = \left[ 2 \pi n(z^*)^2 r_o^2 m_o c^2 / \beta^2 \right] \ln \left( 2 m_o c^2 \beta^2 r_o^2 w_o / I_{\text{adj.}} \right) - \beta^2 - 2(c/z) - \delta \]

(2.6.1)

where \( z^* \) is the effective charge of the ionizing particle; \( n \) is the density of electrons in the detector; \( r_o (= e^2 / m_o c^2) \) is the classical electron radius; \( r = (1-\beta^2)^{-1/2} \); \( I_{\text{adj.}} \) is the mean excitation potential of the material; \( c/z \) is the tight binding shell correlation and \( \delta \) is the correction for the density effect.

The tracks are formed only if the restricted energy loss (REL) rate, \( (\frac{dE}{dX}) \) as given by eq. (2.6.1) is greater than a minimum value \( [(\frac{dE}{dX}) \] \( _{w<w_o} \) crit. depending upon the detector. Therefore, according to this model the condition for track registration takes the form \( (\text{REL}) > (\text{REL}) \) \( _{w<w_o} \) crit. for the material. The restricted energy loss model gives the value of \( [(\frac{dE}{dX}) \] \( _{w<w_o} \) crit. in absolute units unlike the primary ionization model where \( [(\frac{dJ}{dX}) \] \( _{w<w_o} \) crit. does not give any absolute value.
2.6.4 Critical dose of ionization energy criterion

A new criterion for track formation in solid state nuclear track detectors was proposed by Katz and Kobetich in 1968. According to this criterion, at higher energies the principle mode of energy loss is ionization and the tracks are formed through the deposition of energy of secondary electrons along the particle trajectory. They also suggested that tracks in the detector are produced when the critical dose \( D_{\text{crit}} \) of ionization energy is deposited by secondary electrons at a critical distance \( R_{\text{crit}} \) from the particle's path. The special distribution of ionization energy in the vicinity of the incident particle trajectory is a function of particle energy and the energy of secondary electrons. The range energy relation indicates the existence of a critical dose \( D_{\text{crit}} \) to be deposited at a critical distance \( R_{\text{crit}} \) from the particle's trajectory, only above which the tracks will be formed.

2.6.5 Radius restricted energy loss criterion

The Radius Resticted Energy Loss (RREL) criterion is the modified form of Restricted Energy Loss (REL) criterion. It includes the energy deposited in all the events occurring within the radius \( r \) of the particle trajectory. RREL criterion gives central importance to the region near the track whereas REL neglects the ionization events close to the track and gives primary importance to the limiting energy \( w_0 \). An approximate representation of RREL is given
by Paretzke (1977) as

$$RREL = \frac{dE}{dX} \frac{az^2}{\beta^2} \frac{R}{\ln\left(\frac{R}{r}\right) - (1 - \frac{r}{R})} \quad (2.6.2)$$

where $R$ is the maximum track width and $a$ is a constant for the given detector medium. The radial distance $r$ from the particle trajectory is the adjustable parameter in this criterion.

A parameter called Lineal Event Density (LED) is also described by Paretzke. This is the number of primary and secondary ionization and excitation events within a radial distance $r$ from the particle trajectory. Although it is difficult to calculate, the quantity can provide an accurate representation of track formation process.

The overall conclusion that can be drawn from the criterion of all the models and mechanism of track formation by ions of different energies is that tracks are formed by ions at energies for which electronic interactions are the dominant mode of energy loss. Etchable trail is usually not produced at the very end of the ion trajectory where nuclear interactions are prominent and become important.
2.7 METHODOLOGY OF TRACK REVELATION, VISUALIZATION AND THEIR EVALUATION

2.7.1 Track revelation

The fixing and enlarging the image of the latent damage trail formed in SSNTD is known as track revelation. Preferential chemical etching, electrochemical etching, grafting and dyeing and track decoration or precipitation are some common techniques for track revelation in SSNTDs.

i) Preferential chemical etching

For track revelation preferential chemical etching is the most general and widely used method in which a rapid dissolution of the damaged region of the trail cone takes place as compared with the bulk material. This technique is the simplest one. The method was first used by Young (1958) for revelation of the fission fragment tracks in lithium fluoride (LiF). The choice of etching solution, the temperature of the etchant and the time of etching are the critical parameters which must be taken into consideration during etching process. Etching solution should be selected such that the tracks with very small angles are produced (etching efficiency should be maximum) and surface of the detector remains optically transparent. Since the tracks should be enlarged enough to be seen under optical microscope, bulk chemical attack is must. Chemical polishes which tend to round corners are not suitable since they
will obscure etched tracks. Oxidising agents are found to be suitable etchants for a number of polymers because they result in required degradation i.e. breaking down of polymer chains at random along their paths. The change in concentration of the etching solution also results in different responses for various type of polymers. The temperature is most important parameter for etching. For plastics, the most frequently used etchant is the aqueous solution of NaOH or KOH with concentrations typically within the range 1 to 12N. During etching, the temperatures of etchant usually employed are in the range 40 - 70°C. In general, addition of ethyl alcohol to the etchant reduces the threshold value of primary ionization at which tracks become etchable (Somogyi, 1977). However this treatment renders the plastic more brittle and in CR-39 it has been found to reduce the sensitivity of CR-39 (Somogyi and Hunyadi, 1980) for track revelation. When lexan is etched in NaOH, the sodium salt of bisphenol-A anion is precipitated (Paretzke et al., 1973; Gruhn et al., 1980). The presence of high concentration of this etch product accumulated in etchant was found to increase the etching sensitivity of lexan (Peterson, 1970). Frank and Benton (1969) found that the bulk etch rate of Lexan in NaOH solution increased approximately as the square of the etchant normality.

The nature of tracks formed in the detector material is generally characterized by the charge, mass and energy of the
incident particle and also depends on the environmental conditions at the time of irradiation and pre-etching treatments. The shape of a track formed by a charged particle is governed by simultaneous actions of the etchant along the trajectory (damaged region produced by the particle in the detector medium) and on the bulk material. A schematic diagram of chemical etching of a charged particle in a SSNTD is shown in figure 2.3.

For the measurement with high accuracy, long etch times are preferable and fine control of the etching temperature (within ±0.1°C) is often necessary. For the reproducibility of results, the quality of etchant should be controlled carefully and fresh etchants should be replaced to avoid the build-up of the etch products in etchants. To obtain effective etching, constant stirring during etching process and repeated cleaning steps can be adopted to prevent accumulation of etch product layers (Enge et al. 1975; Khan, 1973).

ii) Electrochemical etching

The technique, electrochemical etching of enlarging the damaged trails to sizes, typically up to ~200 μm, gives better application of tracks in the dielectric medium. This was first suggested by Tommasino (1970). In this technique, the irradiated sample is used to separate a cell into two halves containing a suitable etchant. A high electric field (~10-50 kV cm⁻¹) of high frequency i.e.
Figure 2.3: Geometry of track etching
sinusoidal type is applied to the two platinum electrodes inserted in each half of the cell, containing the etchant. Usually, the frequencies in the range of several kHz to hundreds of kHz are employed. Although the theory of the electrical phenomena leading to the break down of the dielectric and the consequent formation of the etch spots is not fully resolved however, it is found that the conducting etchant begins to etch out a cone along the trajectory. Thus an etch pit would be formed along the damaged region by chemical etching. Then the field lines become concentrated at the cone tip and high electric fields are produced in the vicinity of etched track tips leading to sparks to form enlarged tracks. The optimum electrochemical etching (ECE) parameters, viz. electric field strength, frequency and etching conditions have been studied by many workers (Somogyi, 1977; Al-Najjar et al., 1979; Durrani and Al-Najjar, 1980). This method is widely applied to neutron, proton, alpha particle and fission fragment tracks revelation using plastic track detectors.

iii) By track decoration

The method of track decoration was successfully applied to fission tracks by Childs and Slifkin (1962a,b; 1963), in which they employed a combination of pulsed light and electric field to sweep photo electrons into the interior of the damage region of the single crystal. This method was used for visualization of latent tracks of
multipronged events from 1.5 GeV proton interactions, heavy primary cosmic ray particles and alpha particles from polonium. Relativistic C, N and O groups registered observable tracks with this method, but relativistic protons and alpha particles did not give developable tracks. This method was applied by Fleischer and Price (1963a) to decorate the tracks in a glass doped with silver using adequate heat treatment and electric field along the particle trajectory.

2.7.2 Track visualization and evaluation

Commonly used techniques for track visualization are:

i) By electron microscope

The damaged trails can be seen directly by an electron microscope in certain crystalline detector materials such as mica (Silk and Barnes, 1959). The visualization of track can be done either in diffraction contrast mode or thickness contrast mode. In the diffraction contrast mode the crystal planes of the detector are bent such that electrons get scattered out by the Bragg reflection image of the damaged regions which appear as black streaks. In the thickness contrast mode the irradiated sample detector is viewed under an electron microscope after the preliminary etching. This technique can be applied only to extremely thin (< 3000 Å) detector samples with fairly high track density upto $10^5$ tracks/cm$^2$. 
ii) By optical microscope

The optical microscope is the most suitable equipment used for track visualization and counting the tracks in SSNTD having the track density up to $10^5$ tracks/cm$^2$. Tracks produced by preferential chemical etching are usually scanned by an optical microscope at different magnifications. For the purpose of obtaining track densities, the microscope can be focussed on the surface of the detector where the intersection of the track with the surface is seen as a dark circular spot. By little defocussing, one can also look into the depth of the track. For oblique incidence, the track can be seen by changing the focus with the help of microscope and track length can be measured. To obtain the track density or total number of tracks, area of the detector surface is scanned using an eye piece equipped with a grid marked graticule. The calibration of the field of view can be done with the help of a stage micrometer glass slide to find out the scanned area. Counting of the tracks in each graticule can be done with the help of hand tally counter. Track parameters such as track length, track diameter etc can be measured with the help of a micrometer eye piece. By optical microscope, practical visual scanning limit lies between $10^2 - 10^5$ tracks/cm$^2$. This technique is not suitable when a detailed structural analysis of an individual etched channel is desired.
iii) By naked eye

The technique was suggested by Fleischer et al. (1966b) known as the Aluminium backed plastic detector technique in this an opaque coating of aluminium is made one side of the thin detector. The thickness of the detector should be less than the expected length of the particle track. After irradiation, only the detector side is etched. Thus the etchant (hydroxide solution) dissolves the thin layer of aluminium along the track site and produce large holes in it. The holes are easily visible by naked eye.

An alternative technique of detecting holes in thin plastic sheets was also developed by Cross and Tommasino (1967). This technique can be used only upto $10^3$ tracks/cm$^2$. Block et al. (1969) developed another technique in which ammonia gas was made to pass through the track channels from one side of the plastic track detector to form a replica of track pattern on a sensitized paper. It is useful only for very low track densities viz. 1 track/cm$^2$. Dye technique (Khan, 1971) is also employable for track densities less than 10 Tracks/cm$^2$. In this technique a dye is pressed through the etched track holes in a thin detector kept close on a blocking paper to leave the large spots on it which are easily visible to naked eye.

iv) By gas flow or ionic measurement technique

The method proposed by Fleischer and Price (1963a) is important
only for historical point of view. In this method an irradiated specimen of mica serves as a barrier between two halves of a cell containing HF solution and physical measurements such as gas flow or ionic permeability are done for the track counting through the irradiated specimen. The conductivity is measured for several specimens as a function of time and a reproducible curve can be obtained.

v) By image analysis using electronic circuits

The visual counting of etched tracks in SSNTDs by an optical microscope is time consuming. It becomes more tedious when whole area of the exposed track detector is to be scanned such as for absolute fission rate measurements (Azimi-Garakani and Williams, 1977). To overcome this problem, many laboratories have started image analysis using computer controlled electronic circuits. Khan and Durrani (1972) setup an electronic counting system and projection system of etched tracks in SSNTD. Quantimat-900, Magiscan-2, eitz T.A.S. and Reichert-Jung IBAS are commercially available for automatic images analysis. These scanning systems remove the tedious job of counting traditionally associated with the manual and ocular measurements in SSNTDs by enhancing the speed of evaluation. These systems are fairly reliable within the limits given by the optical microscope. These systems require advanced and sophisticated fast computers and thus are very expensive.
2.8 PLASTIC TRACK DETECTORS

Solid state nuclear track detectors have been extensively used for widespread applications in almost all branches of nuclear science and technology. It is gratifying to note that SSNTDs now occupy an essential and sometimes an unique place amongst the techniques and procedures deployed in scientific researches at the very frontiers of knowledge. Out of the SSNTDs the plastics in general are the most sensitive class of materials and are being used with great success to discover new cases of recently reported noble and extremely rare modes, radioactive decay of heavy nuclides (with $Z \geq 88$) involving monoenergetic heavy ions. Now-a-days several types of plastic detectors are known which can record the tracks of charged particles up to relativistic range of heavy ions.

Cellulose nitrate, cellulose acetate, cellulose triacetates, polycarbonates, polyethylene terephthalates etc. of different kinds are some commonly used plastics. It was observed that CR-39 and cellulose nitrate plastics can even record the tracks of protons. Although the polycarbonates (Lexan, Makrofol etc) can not record the etchable tracks of protons, but are capable of recording the tracks of low energy alpha particles and all other heavy ions and fission fragments. Polyethylene terephthalate plastics such as Melinex-0 and Cronar do not record the tracks of alpha particles as well as protons but these are capable to record the etchable tracks of ions.
heavier than $^{11}$B. These threshold characteristics of polyethylene terephthalate and polycarbonate are well understood and can be very usefully utilized for the detection of heavy ions as well as fission fragments without background tracks. As the polycarbonate track detectors such as Makrofol are insensitive to light charged particle $X$ and $\gamma$ rays, they offer a very convenient way of detecting heavy ions in the study of composition of cosmic rays, heavy ion reactions and exploration of super heavy elements etc. (Fleischer et al., 1975; Durrani et al., 1987).

Different types of plastic detectors are available and each of them has unique characteristics for specific applications. The detectors used in the present investigations are described below.

2.8.1 CR-39

Cartwright et al. (1978) were the first to introduce CR-39 polyallyldiglycol carbonate as particle track detectors. CR stands for Columbia Resin. These detectors are found to have some unique properties determined by the chemical and physical structure of CR-39 polycarbonate. CR-39 is a polymer consisting of short polyallyl chains joined by links containing carbonate and diethylene glycol groups into a dense three dimensional network. At the branching point of this network two links consist of polyallyl chains and one consists of diethylene glycol bis allyl carbonate (Stejny, 1987). The molecular structure of CR-39 polycarbonate
The formation of network during polymerization is a special case of percolation process and can be modelled by a Monte Carlo simulation. The model predicts inhomogeneities in cross-linking density which could explain some anomalous etching behaviour of the detector. Polymerization of CR-39 is a transformation from the unsaturated tetrafunctional monomers to the saturated polymer. The process of the formation of a three dimensional network forms various closed zones which contain unused monomer molecules and characterize the residual unsaturation left in the polymer material. The structural inhomogeneities which are responsible for the non-uniformity and non-reproducibility of etching response of CR-39 polymer, is due to isolation of random trapping of unused monomers. To overcome these problems, curing, a process to ensure uniform polymerization, is required. Due to exothermic nature of the polymerization of CR-39 (Turner et al., 1981), specifically
controlled temperature time cycles which were derived assuming a constant rate of polymerization (Somogyi, 1981), were employed during polymerization. Fowler et al. (1979) introduced controlled 'time-temperature' curing cycles for the fabrication of CR-39 detectors. Before the polymerization is complete, the system enters a glassy state. The residual mobility in glassy state and presence of chemically reactive groups are responsible for physio-chemical aging of the polymer and the changes in the performance of CR-39 with time.

CR-39 has similarities with glass in its optical properties. It has absolute clarity and abrasion resistance similar to glass and also environmentally very stable. It has sp.gr. 1.32 at 25°C and refractive index 1.504 at 25°C and also has same light transmission as glass (92% for 0.25" thickness, Pershore Moulding’s data sheet, PM-002).

The high sensitivity of CR-39 to charged particles was discovered by Cartwright et al. (1978). Following this discovery Hayashi and Doke (1980) reported that their CR-39, made by Sola Optical, Japan using PPG monomer could record relativistic carbon nuclei (z/β = 6) in primary cosmic radiation. Now this plastic has been successfully applied to record nuclear charged particles of z/β value from 6 to 100 (Manjoor et al. 1988). CR-39 is remarkably
sensitive to charged particles, even at relativistic velocities to heavy ions with z as low as 18 (Ar) and up to ~ 60 MeV in the case of α-particles (Cartwright et al., 1978). For precise and sensitive applications on a large scale CR-39 polycarbonate has to be made highly sensitive with consistent and reproducible track recording characteristics. The sensitivity, resolution and optical properties of CR-39 are unrivalled among track recording solids. Even after dissolving 500 μm thickness by etching, the surface remains glassy smooth. Since the discovery of the unique sensitivity of CR-39 among the plastics to ionizing particles, intense efforts have been made to investigate its properties and to develop its practical applications in numerous fields. CR-39 is insensitive to γ and X-rays and is sensitive to both light and heavy charged particles, it is widely used in heavy ion research for example composition of cosmic rays, heavy ion nuclear reactions, radiation dose due to heavy particles, explorations of extra heavy elements and magnetic monopoles etc (Durrani and Bull, 1987). It also has progressed to a leading position owing to its transparency to visible light and small value of critical losses.

2.8.2 CR-39 (DOP)

CR-39 (DOP) is a modified product of CR-39 manufactured by Pershore Molding (U.K.). The sensitivity of CR-39 might be affected by factors such as the purity of the monomer, the initiator concentration and the temperature during the polymerization (Fowler
et al., 1979; Price et al., 1979; Price and O'Sullivan, 1981). The addition of small amounts of certain chemical additives to CR-39 will affect changes in its sensitivity, response and surface etching properties, although the mechanisms underlying these changes have not been well understood. The optical and etching properties of CR-39 can be improved by incorporating additives such as dioctyl phthalate (DOP) in the polymer (Price and O'Sullivan, 1981; Fujii and Yokota, 1986). A heavy phthalic acid ester such as DOP acts as a plasticizer and that CR-39 with DOP has good etching properties. A plasticizer increases the free volume of a polymer, producing the polymer chains more mobile. A good plasticizer needs to be soluble in the monomer/polymer and should have sufficiently low molecular weight to increase the free volume without being low enough to diffuse out of the plastic (Portwood and Stejny, 1984). Some plasticizers have been used to decrease or completely eliminate the coalescence of the post etched surfaces. Several phthalate esters commonly used in polymer technology as plasticizers have been tested in CR-39 plastic (Price and O'Sullivan, 1981). There could be either a decrease or increase in the sensitivity of CR-39 (DOP) compared with that of CR-39 (no additives) depending upon the concentration of additives (Anupam et al., 1991). It was also reported by Tarle (1981) that a heavy phthalic acid ester such as DBP or DOP acts as a plasticizer making a more homogeneous polymer and that CR-39 doped with DOP has better etching properties than CR-39.
The reduced sensitivity for CR-39 doped with DOP may be due to the presence of aromatic rings in DOP. Aromatic ring acts as an electron sink and hence will tend to decrease the sensitivity of the polymer to charged particles. After doping, a remarkable improvement in etching homogeneity and optical transparency of the post etch surface of detectors were observed even after using long etching periods. CR-39 polymer exhibits surface opaqueness after prolong etching in hot aqueous solution. CR-39 (DOP) polymer is free from this limiting property of CR-39.

2.8.3 Makrofol polycarbonate

Makrofol polycarbonates manufactured by a casting process into the form of thin sheets by Bayer AG of Leverkusen, West Germany, have same composition \( \text{C}_{16}\text{H}_{14}\text{O}_3 \) as Lexan, a polycarbonate manufactured by General Electric Co. of USA. However these polycarbonates have different type of behaviour than Lexan polycarbonate. Different types of Makrofol polycarbonates such as Makrofol-KG, KL, E and N etc. produced by different manufacturing processes are expected to behave in different ways (Illari et al., 1977). Out of these Makrofol-KL contains a slight amount of a colour dye while Makrofol-N is a trade name of yellow polycarbonate.

Makrofol polycarbonate plastics (much useful as heavy ion as well as fission track detectors) were originally used as insulators in electrical devices. Makrofol-E and N are sensitive to particles
having $z > 2$ and $z > 8$ respectively (Todorovic, 1990). Makrofol KG, KL and N are not sensitive to α-particles and other lighter charged particles. The shape of tracks produced by heavy ions and fission fragments are needle like with a slight spread towards its tail.

During the last decade thin sheets of polycarbonates have been used to produce microfilters and single pore membranes for their remarkable applications in the field of environmental (Fischer and Spohr, 1983 and references therein), bio-medical (Roggenkamp et al., 1981) and superfluidity (Gamota, 1973). Makrofol polycarbonates also offer a very convenient way of detecting heavy ions in the study of composition of cosmic rays, heavy ion nuclear reactions and exploration of super heavy elements etc. because these are insensitive to light charged particles, X and γ-rays (Fleischer et al. 1975; Durrani et al., 1987).

2.8.4 Lexan polycarbonate

Lexan (bisphenol-A polycarbonate) has the chemical composition $C_{16}H_{14}O_3$ and is manufactured by the General Electric Plastics Dept., Mt. Vernon, Indiana, USA. It has been the most widely used plastic for the detection of fission fragments as well as for particle identification, with a greater variety of etch pits having consequently been utilized. A charge resolution $\Delta z$ of 0.3 charge units and a mass resolution of ~ 2 amu in Lexan for cosmic rays in
the region of $12 \leq Z \leq 30$ were obtained by Price et al. (1970). Paretzke et al. (1973) have examined the etching reaction of Lexan with NaOH and the reaction mechanism was found to be

$$\text{[} \begin{array}{c} \text{CH}_3 \\ \text{O} \\ \text{CH}_3 \\ \text{O} \\ \text{C} \\ \text{O} \\ \text{C} \end{array} \text{]} + 4 \text{OH}^- \rightarrow \text{[} \begin{array}{c} \text{CH}_3 \\ \text{O} \\ \text{CH}_3 \\ \text{O} \\ \text{C} \\ \text{O} \\ \text{C} \end{array} \text{]} \ + \text{CO}_3^{2-} + 2\text{H}_2\text{O}$$

The bisphenol-A anion is released from the polymer chain.
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