CHAPTER 2

TECHNIQUES, MATERIALS AND METHODS
Chapter-II

2.1 Radiation Detection

Detector is the device which senses the interactions of radiations and converts the interaction to measurable signal. Radiation detection techniques are based on measurement of the charge produced due to interaction of radiation as it passes through detector volume. The net result of the radiation interaction is the appearance of an electric charge within the active volume of the detector (Evans, 1978). The interaction times are so short (Friedlander et al., 1981) that the deposition of radiation energy can be considered as instantaneous and charge appears in the detector at zero time. This charge which is the basic signal for radiation detection must be collected. Charge is collected by applying an electric field across the detector. Time required for charge collection greatly varies from one detector to another depending on the mobility of charge carriers in the detector and the average distance to be traveled.

2.2. Solid State Nuclear track detectors (SSNTDs)

Solid State Nuclear track detectors (SSNTDs) have been employed for recording the charged particle tracks. These detectors have been used for long time for radon, thoron and radon exhalation rate measurements. When energetic charged particles pass through a dielectric medium, they leave a trail of damaged molecules along its track. This is similar to the track recorded by charged particles in nuclear emulsions. Inorganic insulators like quartz, mica, glasses and zircon as well as organic polymers like Lexan, CR-39 and LR-115 type II are widely used as Solid State Nuclear Track Detectors (SSNTDs). Initial track produced by radiation is very small (a few nm) and can be seen only under an electron microscope. However, etching the detector in alkali or acid tracks can be developed on etching the damaged tracks are preferentially etched out to form small (10-20μm) pits that can be easily seen and counted under a microscope. The sizes of the pits made by alpha particles are substantially small as compared to the pits made by fission fragments. These SSNTDs are insensitive to fast electrons, gamma rays and
even protons. Visible tracks in different materials can only be produced if the specific energy loss \((-dE/dx)\) is above the minimum threshold value for that material.

2.3 Track Formation Criteria

When a heavily ionizing nuclear charged particle passes through an insulating solid, the physical, chemical and other properties of the solid along and around the path of the particle change and a narrow path of intense damage is created. This narrow path is called latent track and the track recording materials are known as solid state nuclear track detectors. Various authors have suggested to relate the track formation with different parameters, such as total rate of energy loss, primary ionization, restricted energy loss etc. The experimentally set up track formation criterion is that there exists a certain value of energy loss rate known as critical value of energy loss rate below which the tracks are not formed in solids. The critical energy loss is generally denoted by \(J_c = (dE/dx)_c\). The values of which is different for different solids but same in a given solid for all the particles as shown in Figure-1. It can be seen from this figure that particle may not be able to produce track if either energy is too low or too high. However if the value of \(J\) is less than \(J_c\) for a given solid, the tracks will not be formed in the solid at any energy of the traversing particle.

The charged particle latent tracks are narrow (< 50Å radius) and stable chemically reactive centers of strain, composed mainly of displaced atoms rather than electrons (Maurette, 1970; Fleischer et al., 1975).
Figure-1: Curves of primary ionization rate as a function of relative velocity ($\beta$) and energy per nucleon for a number of ions. The experimental points for accelerator ions in lexon polycarbonate are given as open circles for zero registration and as filled circles for 100% registration. Thresholds for other detectors are also indicated.
On the basis of experimental tests conducted (i) on the measurement of effects of electron irradiation on chemical dissolution rates of solids and (ii) on the measurement of radial distribution of etchable damage in solids, it has been concluded that two separate mechanisms of track formation exist, one for the inorganic solids and glasses and the other for organic solids or polymers. In case of inorganic solids and glasses, the ion explosion spike model proposed by Fleischer et al., (1965) provides a considerably satisfactory explanation of track formation mechanism. According to this model, a positively charged particle knocks out the orbital electrons of the atom lying in and around the vicinity of its path, thus producing the cylindrical region full of positive ions. These positive ions thereupon repel one another violently, thus disturbing and distorting the regular lattice in a crystalline solid (Figure-2) and producing a more or less cylindrical region (Bonfiglioli et al., 1961; Lindhard and Thomson, 1962; Chadderton and Montagu Pollock, 1963; Benton, 1967; Fleischer et al., 1967). On the other hand, in case of organic polymers and plastics it is believed that both the primary and secondary ionization and excitation play an important role in the production of etchable tracks in the organic polymers. These broken molecular chains rarely unite at the same place, instead they produce broken bonds and free radicals etc. which are chemically more reactive as shown in Figure-3. These latent damage track trails can be developed by using the appropriate etchant and can be visualized then under the optical microscope. It is explained as follows: as the damage trails are chemically more reactive regions (compared to the undamaged area), on increasing the sample in appropriate etchant, all those trails which intersect the surface are rapidly dissolved and hollow cylindrical tubes about 50 Å diameter are left behind. The undamaged well of hollow tube is attacked and its diameter increases into micron range, thus ultimately, a modified, enlarged version of the original damage trail is produced which can be seen easily under an optical microscope.
Figure-2. The ion explosion spike mechanism for track formation in inorganic solids
Figure 3. Chain cleavage and track formation in organic polymers.
The radiation damage trails are more susceptible to chemical reaction as compared to other bulk materials because of the large free energy associated with the disordered structure. The technique of enlarging the latent trails of radiation damage with suitable chemical agent is called chemical etching. The shape of the etched track in certain materials depend 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 irradiations and pre-etching treatments. The shape of the track also depends on the ratio of chemical attack along the track $V_T$ to the rate of chemical attack for the bulk material $V_B$. This process of track revelation involves the concept of critical angle given as

$$\theta_c = \sin^{-1}(V_B/V_T)$$

If the angle of incidence of the charged particles with respect to the surface of the detector is less than the critical angle, the tracks of the incident particles can not be revealed (Fleischer et al., 1975). As the etchant moves in the material along the damage trails at a faster rate, i.e. $V_T > V_B$, it produces a conical etch pit as shown in Figure- 4. The etching efficiency of the detection depends upon the critical angle and is given by

$$\eta = 1 - \sin \theta_c$$

The etch rate depends up on the composition of the material, the nature of the the damage produced by the charged particle, etchant parameters and etching time.
Figure- 4. Track-etching geometry. Preferential etching along tracks at a rate $V_T$ plus general etching at a bulk rate $V_B$ produce conical holes with a cone angle that can be shown to be $\sin^{-1}(V_B/V_T)$. The angle is larger and the pit is shallower for the less intense track on the right.

(A) shows the unetched sample

(B) and (C) shows the progressive effects of etching
2.4 Radon Measurement Techniques

There are many techniques for measuring the concentration of radon in soil, air, groundwater and dwellings. The techniques are based on the detection of emissions from radioactive decay of radon and its daughter products. Most of the methods are based on the detection of alpha particles, some on detection of beta emission while a few utilize gamma decays. The techniques of radon measurement may be divided in two categories.

(i) Instantaneous radon measurement techniques

(ii) Time integrated long term radon measurement techniques

2.5 Instantaneous radon measurement techniques

2.5.1 Grab Sampling Technique

This technique gives an instantaneous value of the radon concentration in air sample taken in a container for the analysis (Lucas, 1957; George, 1976). Inside the dwelling, the radon daughter products are measured by collecting airborne particulate on the filter paper for a period of 2 to 10 minutes (Raabe and Wrenn, 1969; Rolle, 1972). The filter paper sample is obtained by sucking air through a 2.5 cm diameter millipore filter type of 0.8 μm pore size at the suction rate of 20 lpm for a time period ranging from half an hour to one hour. Due to short half-lives of the radon decay products, the analyses are performed strictly within one hour after the sampling. The filter paper sample is counted using a ZnS (Ag) detector system, which is coupled to a nuclear counting system to record the counts due to disintegration of radon daughter collected on the filter paper. Grab sampling technique is suitable for large-scale surveys, because of its simplicity, minimal labor requirement and low cost. The disadvantage of this technique is that it does not give accurate information on time-averaged air concentration in dwellings.

2.5.2 Ionization Chamber Technique

This is a gas filled electrode system, which is designed to detect the presence of an ionizing particle (Wrenn et al., 1975). The ionizing particle creates a pair of positive and negative ions when the gas passes through the chamber. These ions are attracted in
the opposite direction under the influence of electric field. The ionization chamber is operated in the range of potential varying from 100 to 300 volts so that the multiplication and recombination of the opposite pair is negligible. Ionization chamber technique can be used for measuring radon concentration down to 10 Bq m\(^{-3}\) (Pacer and Czarnecki, 1980).

### 2.5.3 Radon Emanometry

This technique is used to measure the alpha emanation rate from radon in the gas fraction of a soil or water sample by pumping the gas into a scintillation chamber using a closed circuit technique. Emanometer was the earliest device to measure radon and is still widely used for quick radon survey (Cheng and Poritt, 1981; Ramola et al., 1989; Virk, 1990).

### 2.6 Time integrated long term radon measurement techniques

Time-integrated schemes involve the accumulation of radon over longer time periods from a few days to a week or more. In these techniques, the radon is measured either directly by detecting the alpha emission or indirectly by detecting the radioactive decay products of radon. Brief description of these techniques is given below:

#### 2.6.1 Charcoal Technique

In this technique radon can be indirectly measured by the determination of radon decay products present in the sample (Cohen and Cohen, 1983). A charcoal sampler with dimensions of the order of 10 cm can be used to collect 222Rn over a period of a day to a week. The instrument consists of a small container filled with activated charcoal. Gamma rays emitted from \(^{214}\)Pb and \(^{214}\)Bi which are daughter products of radium, are measured. With a lower detection limit of 0.2 pCi/L for sixty hours of exposure, this instrument is suitable for measuring indoor radon levels. This technique is helpful for a period of more than a week, because after this time period most of the \(^{222}\)Rn collected at the beginning will decay out due to its 3.82 days half life period.
2.6.2 Plastic Bag Technique

Plastic bag technique is also the time-integrated technique for radon (Sill, 1969) measurement. The sampled air collected for a period of two to three days is pumped into a bag which is impermeable to radon gas. For alpha counting, the integrated sample during analysis is transferred to the Scintillation flask. Concentration as low as 0.01 pCi/L can be measured using this technique.

2.6.3 Thermo luminescence Detector Technique

This is a process in which light is emitted by a substance when it is heated and which can be attributed to the previous exposure to ionizing radiation. When a crystal is exposed to ionizing radiation, electron hole ionization pairs are created and some of them combine with the trapped charges. Thermoluminescence detectors, used for radon measurement are very thin wafers (760 m of calcium sulphate doped with dysprosium in a matrix of Teflon), which are mainly sensitive to alpha radiation (McCurdy et al., 1969; Pacer and Czarnecki, 1980). These detectors are typically exposed for 30 days in the uranium exploration program and then processed for their thermoluminescence peaks.

2.6.4 Alpha Meter Technique

This technique employs a solid-state alpha particle detector counter assembly. The alpha particle detector is a silicon-diffused junction with an active area of 400 mm². When an alpha particle enters the n-p junction, a number of electron hole pairs are generated which are proportional to the energy of alpha particle. The flow of current is sensed and amplified many hundreds of times to produce a pulse that may then be counted. The alpha counts are displayed on the light emitting diode (LED) display (Warren, 1977). The other instrumental method of measuring the radon flux includes the alpha cards with a central collector for recording radon daughters (Dyck et al., 1983). The detector records the alpha particles emanated by radon isotopes and their alpha emitting daughters. This instrument does not register any alpha particle having energy less than 1 MeV.
2.6.5 Track Etch Technique

Track etch technique is one of the most widely used techniques for radon measurement (Klies et al., 1992; Bhagwat, 1993; Ramchandran, 1998; Kumar et al., 2003; Mahur et al., 2006). In this technique, the monitoring device is a solid state nuclear track detector (SSNTD). The principle of detection consists of the damage imparted in the detector material by alpha particles from radon and its decay products. The damage imparted in the detector is observed under optical microscope. Heavily ionizing particles passing through insulating media leave narrow trails of damage on an atomic scale (~30-100 Å). With each alpha particle producing a distinguishable track, these latent tracks can be enlarged to microscopically visible size by the method of chemical etching in which the damaged region reacts at faster rate with a chemical reagent. Number of tracks per unit area in the detector is proportional to the average exposure rate and exposure time. Exposure time can range up to a year or more, if desired, using improved plastic track detectors which retain alpha tracks without fading for a very long time at ambient temperatures. Though several detector materials have been developed but LR-115 and CR-39 are the two most popular track detectors used for radon dosimetry. The advantage of track etch technique is its simplicity, no involvement of electronics, low cost and feasibility in extensive radon surveys. A strong drawback in the use of solid-state nuclear track detectors is that they only integrate the received flux of particles and don’t provide time dependent response. Other demerit of track etch technique is its poor sensitivity for integrated time periods less than one month. In studies of indoor radon, monitoring time period of about three months is adequate.

Track etch technique was used in the present study. In this technique LR-115 type II track etch detector was employed for measuring the Potential Alpha Energy Concentration (PAEC) of radon progeny in Working Level (WL) units. LR-115 type- II films are sensitive to α particles of energy range 0.1 to 4 MeV and are unaffected by electrons, X-rays and γ-rays (Jonsson, 1981). Due to its being sensitive to only a specific range of energy, it is ideal for the use as radon dosimeter even in bare mode as it is free from plate out effect (Abu-Jarad et al., 1980).

The pieces of the detector film of size 2 × 2 cm, fixed on a thick flat card are exposed in “Bare mode” by hanging the cards on the wall in the room for a period of few months such that the detector views a hemisphere of radius at least 6.9 cm, the range of
$^{214}\text{Po}$ α-particles in the air. No surface should be closer than this range as the decay products would act as an indeterminate α-particles sources. Detectors are mounted vertical and the locations are so selected that the dust collection on the detectors be minimum. After exposure, the latent tracks due to radiation damage produced by alpha particles from radon and its progeny are revealed by chemical etching in a constant temperature water bath (Figure-4). The counting of alpha tracks are done using a binocular optical research microscope as shown in Figure-5.
Figure 4: Constant temperature water bath
Figure-5 Optical binocular optical research microscope with a magnification of $400 \times$
2.7 Radon and Thoron measurements using twin chamber dosimeters

Twin cup radon dosimeter was used in present study for the measurement of indoor and outdoor radon concentrations. The alpha sensitive plastic track detector, pelliculable LR-115 Type II (Manufactured by Kodak Pathe, France) is used. It is a 12 μm thick film red dyed cellulose nitrate emulsion coated on inert polyester base of 100 μm thickness and has maximum sensitivity for alpha particles, fission fragments and ionizing particles with high enough LET. It is widely used for particle detection of ionizing particles, high-resolution neutron radiographic uses, alpha radiography, cosmic ray investigations etc. The film can be used to record the tracks of protons with an energy <100 KeV and alpha particles with energy ~0.06 to 6 MeV. It is insensitive to X or γ-rays, photons, electrons and high-energy protons. The sensitivity is claimed to be one of the best amongst any other plastic detectors. For fast neutrons, it has low detection efficiency (10⁻⁵ track/neutron) (Khan, 1975). Track etching mechanism of pelliculable LR-115 has been studied at different temperatures ranging from 30 °C to 60 °C for different etching times and the calculated value of activation energy is 0.1845 eV (Paul and Bose, 1980). The recommended etching conditions given by the manufacturer are 2.5N NaOH, 60°C, 65 to 95 minutes without agitation. Another suitable etching condition reported is 2.5N NaOH, 60°C, 60 to 70 minutes with stirring (Costa-Riberio and Labao, 1975).

The dosimeter has been developed at Bhabha Atomic Research Centre (BARC) and is shown in Figure- 6. Each cylindrical chamber has a length of 4.1 cm and a radius of 3.1 cm. One piece of the detector film (SSNTD) placed in compartment M measures radon only which diffuses into it from the ambient air through a semi-permeable membrane. It allows the build up of about 90% of radon gas in the compartment and suppresses thoron gas concentration by more than 99%. The mean time for radon to reach the steady-state concentration inside the cup is about 4.5 hour. Glass fiber filter paper in the compartment F allows both radon and thoron gases to diffuse in and hence the tracks on second piece of the detector film placed in this chamber are related to the concentrations of both the gases.
Third piece of the detector film exposed in bare mode (placed on the outer surface of the dosimeter) registers alpha tracks attributed to both the gases and their alpha-emitting progeny, namely $^{218}\text{Po}$, $^{214}\text{Po}$, $^{216}\text{Po}$, and $^{212}\text{Po}$. All the detectors are exposed for 90-100 days. After exposure the films are etched. The detectors are pre-sparked using spark counter as shown in Figure-7 (Cross and Tommasino, 1970) to fully develop the partially etched track holes. The tracks are then counted at the voltage corresponding to the plateau region of the counter. The concentrations are derived from the observed track densities using appropriate calibration factors. The calibration factors depend upon various parameters such as membrane and filter characteristics as well as on the energy of the alpha particles for the cup mode exposure, the parameters of the etching process and spark counting characteristics.
Figure -6 Twin cup radon-thoron dosimeter
Fig-7 Spark counter set up
2.8 Measurement of radon exhalation rate

Radon exhalation rate is of prime importance for the estimation of radiation risk from various materials. "Sealed Can Technique" (Fleischer and Morgo-campero, 1978; Mahur et al., 2008) is widely used for radon exhalation measurements in solid samples. SSNTD is fixed on the top inside the cylindrical "Can" as shown in Figure-8. Sample of known amount is placed at the base of the Can. The Cans are sealed for 90-100 days. Thus the lower sensitive part of the detector is exposed freely to the emergent radon from the sample in the Can so that it could record the tracks of alpha particles resulting from the decay of radon in the remaining volume of the Can and from $^{218}$Po deposited on the inner walls of the Can. Radon and its daughters reach equilibrium in about four hours and hence the equilibrium activity of emergent radon can be obtained from the geometry of the Can and the time of exposure. For the study of the radon exhalation rates from building and construction materials like bricks and flooring materials like mosaic, marble, granite, ceramic tile etc., plastic Can of known dimension is sealed by plasticin to the individual building material. In each Can a SSNTD is fixed at the top inside the Can (Figure-9).

After the exposure the detectors are etched in suitable etchants in constant temperature water bath for revelation of tracks. The resulting $\alpha$- particle tracks are counted using an optical microscope. The activity is obtained from the track density in the etched detectors using the calibration factor obtained from calibration experiment. Following expression gives the exhalation rate (Khan et al., 1992, Fleischer and Morgo-campero, 1978 and Mahur et al., 2008):

$$Ex = \frac{CV\lambda}{A \left[ T + \frac{1}{\lambda} \left( e^{-\lambda T} - 1 \right) \right]}$$  \hspace{1cm} (1)

where $Ex$ is the Radon exhalation rate (Bq m$^{-2}$ h$^{-1}$); $C$, the integrated radon exposure as measured by LR-115 type II plastic track detector (Bq m$^{-3}$ h$^{-1}$); $V$, the effective volume of the Can (m$^3$); $\lambda$, the decay constant for radon (h$^{-1}$); $T$, the exposure time (h) and $A$ is the area covered by the Can (m$^2$).
Figure 8. Experimental set up for the measurement of radon exhalation rate using "Sealed Can Technique"
Figure 9: Assembly for the measurement of radon exhalation rate using “Can technique”
2.9 Natural radioactivity measurements

There are many techniques for the measurement of natural radioactivity in the samples. We present three techniques used in the present study for estimation of the natural radionuclides, uranium ($^{238}\rm{U}$), thorium ($^{232}\rm{Th}$) and potassium ($^{40}\rm{K}$) in coal–fly ash, rocks, soil, sand and building construction material samples.

2.9.1 Gamma-ray spectroscopy technique using NaI (Tl) detector

A low level gamma ray spectrometric set up at National Geophysical Research Institute, Hyderabad, was used for estimation of the natural radionuclides, uranium ($^{238}\rm{U}$), thorium ($^{232}\rm{Th}$) and potassium ($^{40}\rm{K}$) in coal and fly ash samples. About 250 g of each sample was packed in plastic containers and sealed for four weeks (El-Arabi, 2005) for $^{238}\rm{U}$ and $^{232}\rm{Th}$ to attain secular equilibrium with their decay products and to prevent $^{222}\rm{Rn}$ and $^{220}\rm{Rn}$ loss. The detector consisted of a 5" x 6" NaI (Tl) crystal coupled to a 5" diameter photomultiplier tube. A 256-channel data set covering 0-3 MeV is obtained through a Multi Channel Analyzer (MCA) card (on a computer) that provided a gain-stabilized spectrum. $^{238}\rm{U}$ and $^{232}\rm{Th}$ were assessed from the intensity of the gamma ray peaks of their daughters, whereas $^{40}\rm{K}$ was measured directly from its own gamma ray of 1.46 MeV.

2.9.2 Gamma-ray spectroscopy technique using HPGe detector

Gamma ray spectrometric measurements were carried out at Inter-University Accelerator Centre, New Delhi using a coaxial n-type HPGe detector (EG&G, ORTEC, Oak Ridge, USA) for estimation of the natural radionuclides, uranium ($^{238}\rm{U}$), thorium ($^{232}\rm{Th}$) and potassium ($^{40}\rm{K}$) in rock and soil samples. Photograph of gamma ray spectrometric set is shown in Figure-10.

Before measurements, the containers having the samples were kept sealed for about 4 weeks in order to reach equilibrium of the $^{238}\rm{U}$ and $^{232}\rm{Th}$ and their respective progenies. The samples were then subjected to high resolution gamma spectroscopic analysis. HPGe detector (EG&G, ORTEC, Oak Ridge, USA) having a resolution of 2.0 keV at 1332 keV and a relative efficiency of 20% was placed in 4" shield of lead bricks.
on all sides to reduce the background radiation from building materials and cosmic rays (Kumar et al, 2001). The detector was coupled to a PC based 4K multi channel analyzer and an ADC for data acquisition. For activity measurements the samples were counted for a period of 72000 seconds. The activity concentration of $^{40}\text{K}$ ($C_K$) was measured directly by its own gamma ray of 1461 keV. As $^{238}\text{U}$ and $^{232}\text{Th}$ are not directly gamma emitters, their activity concentrations ($C_U$ and $C_{Th}$) were measured through gamma rays of their decay products. The spectra were analyzed using the locally developed software “CANDLE (Collection and Analysis of Nuclear Data using Linux Net work)” an example shown in Figure-11.

2.10 Fission track registration technique

For the uranium analyses in some soil, fly ash and building material samples fission track registration technique was used. The method is highly reliable, sensitive and simple. At the same time it has the potential capability of micro mapping even for sub ppb levels of fissionable impurity. The fission fragments resulting from (n, f) reaction on $^{235}\text{U}$ are observed by track etch detectors kept in contact with the sample or standard material. From the number of tracks in the detector, the uranium concentrations in the samples can be determined (Singh et al., 1998). Thermal neutrons for producing fission are obtained from a nuclear reactor.

All samples were grinded properly and sieved through 100 mesh. A homogenous mixture of accurately weighed sample powder and methyl cellulose in the ratio 1:2 ratio by weight was made. 200 mg of this mixture a thin pellet of about 1mm thick and 1.3 cm in diameter was prepared by a hydraulic pellet machine applying a pressure of about 5 t/cm$^2$. Pellets of standard glass of known uranium concentration (Azam and Prasad, 1989) were also prepared in the similar manner. Each of these pellets was sandwiched between a pair of Makrofol –KG plastic track detector pieces of same size. All the pellets of sample and standard glass were enclosed in an aluminum capsule and sealed tightly to make the intimate contact. This assembly was irradiated with a thermal neutron fluence of about $10^{15}$ cm$^{-2}$ (nvt) at APSARA reactor, Bhabha Atomic Research Centre Mumbai, India. After irradiation the detector pieces were separated from the pellets, and etched in 6.25 N KOH solution at 60$^0$ for 20 minutes in a constant temperature water bath to reveal the fission tracks.
Fig. 10  Gamma ray spectrometric set at Inter-University Accelerator Centre, New Delhi
Figure-11 An example of gamma ray spectrum
References


