CHAPTER III

EXPERIMENTAL PROCEDURE
AND WORKING FORMULAE
3.1. Introduction:

The estimation of radon and thoron concentration in dwellings and radium content and radon exhalation rates from soil samples collected from different areas has been done by “particle track analysis” method using SSNTDs. A brief description of the various types of materials and instruments used in the present study and also the various stages of the experimental procedure are given below.

3.2.1. Solid State Nuclear Track Detectors:

In the present study track detectors like LR-115 (type-II) have been used as passive detectors of alpha particles from radon, thoron and their daughters in indoor air and from soil.

Detectors like LR-115 used for indoor radon study and soil radon study are durable, simple, stable and not fogged by exposure to sun light. Fading of the damaged trajectory is also not observed even with moderate heating. These advantages make SSNTDs suitable for studies of environmental radiations like cosmic rays in air craft, alpha exposures due to $^{222}\text{Rn}$ and $^{220}\text{Rn}$ in indoors and in soil as compared to other devices. Characteristics of the alpha sensitive track detectors used in radon dosimetry and their threshold limit of detection are given in Table 3.1 and Table 3.2.

Track density registered on SSNTDs depend on the properties of the detector material and the characteristics of the environment besides the etching conditions.
Table 3.1: Characteristics of detectors used in $^{222}\text{Rn}$ dosimetry

<table>
<thead>
<tr>
<th>Detector Composition</th>
<th>Density (gm/cc)</th>
<th>Trade Name</th>
<th>Proposed Etching Condition</th>
<th>$V_g$ ($\mu$m/h)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cellulose Nitrate ($\text{C}_6\text{H}_8\text{O}_9\text{N}_2$)</td>
<td>1.52</td>
<td>CN85</td>
<td>2.5 N NaOH, 60°C for 90 min.</td>
<td>3.2</td>
</tr>
</tbody>
</table>

Table 3.2: Typical limits of detectable alpha-particle energies for these detector materials

<table>
<thead>
<tr>
<th>Type of detector</th>
<th>$E$ (Min) in MeV</th>
<th>$E$ (Max) in MeV</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cellulose Nitrate ($\text{C}_6\text{H}_8\text{O}_9\text{N}_2$)</td>
<td>0.1</td>
<td>4 - 6</td>
</tr>
</tbody>
</table>

It is necessary to calibrate the system before its use in the field because the sensitivity of the detector material used depends on the dosimeter material and its design besides its variations with time and storage.

The cellulose nitrate ($\text{C}_6\text{H}_8\text{O}_9\text{N}_2$) LR-115 type-II (strippable) film, manufactured by Kodak, Pathe, France has been used for the present study. LR-115 can easily register alpha particle tracks, which can be made observable by chemical etching. The type-II film is thin enough for the penetration of majority of the incident alpha particles to form perforated tracks and also thick enough to make well etched visible tracks.
3.2.2. Dosimeter Cup: (For Indoor Radon Study)

Actual design of SSNTD based dosimeters is based on parameters like

i) the thoron discrimination,

ii) need for aerosols or particulates,

iii) time of exposure etc.

Such type of dosimeters are classified into three categories viz.,

i) diffusion sampler,

ii) permeation sampler and

iii) bare detector dosimeter.

Diffusion sampler is a tube type dosimeter with the detector placed at one end. Other end of the tube is open. Length of the tube is made to allow complete thoron decay during the diffusion to the effective zone area near the detector. Portability and possibility of easy handling of it as a personal dosimeter are the attractive features of this arrangement. This is suitable only for long term measurements.

Permeation sampler differs from the diffusion sampler. In this type of dosimeter, the open end of the tube is closed by a semi permeable membrane that delays the entry of thoron and discriminates between $^{222}$Rn and thoron.$^4$

Bare detector mode used in the measurements consists of a detector affixed on to a rectangular card which is facing the atmosphere to be monitored. This mode of exposure is generally suspected to suffer from interference due to dust deposition on the film. It has been observed that dust loading above 0.30 mg cm$^{-2}$ for a period of exposure of 90 days could affect $^{222}$Rn progenies.$^5$ Different types of $^{222}$Rn dosimeters used by different workers is shown in a tabular form (Table3.4). While designing the dosimeter, the following points are taken in to consideration viz.

i) dosimeter cup,

ii) detector position,

iii) alpha energy range and
Different types of dosimeters used for Rn\textsuperscript{222}/Thoron and their progeny measurements are shown in table 3.3.

### Table 3.3: Different types of Dosimeters used for Rn\textsuperscript{222} /Thoron and their progeny measurements

<table>
<thead>
<tr>
<th>Dosimeter</th>
<th>Dimensions</th>
<th>Uses</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radon dosimeter</td>
<td>Internal diameter of 5.9 cm and a volume of 100 ml with open mouth covered with a suitable semi permeable membrane</td>
<td>\textsuperscript{222}Rn only</td>
</tr>
<tr>
<td>Twin chamber (TW)</td>
<td>The unit comprises a plastic cylindrical vessel open at both the ends. There is a plastic dividing wall in the middle, which divides the whole cylinder into two chambers. Detectors can be attached on both sides of this wall. Detectors can also be attached to the outer surface of the cylinder in a fixed position and exposed to radiation. The open ends are covered by perforated sheets. This type of dosimeter cups are used in the present study. Therefore, details of the dosimeter is discussed in this chapter.</td>
<td>\textsuperscript{222}Rn/thoron gas along with their daughters.</td>
</tr>
<tr>
<td>Bare detector</td>
<td>It is a rectangular type one with the detector affixed on it.</td>
<td>\textsuperscript{222}Rn daughters</td>
</tr>
</tbody>
</table>

#### 3.2.2.1. Details about Plastic Twin Chamber Dosimeter Cup Method Used in the Present Study:

In the present study Plastic Twin-Chamber dosimeter cups (BARC-type) were used to determine the concentrations of indoor radon, thoron and their progenies. The detectors were exposed in three different modes: 1) bare mode, 2) cup with filter
paper, and 3) cup with filter paper and mylar. The preview and schematic diagram of a Plastic Twin-Chamber dosimeter cup with detector positions is shown in Fig. 3.1 and 3.2.

It is a plastic cylindrical vessel of 11 cm. length and 7 cm. diameter and opened at both ends. There is a plastic dividing wall at the middle, which divides the whole cylinder into two chambers each of length 5.5 cm. Detectors can be attached on both sides of this wall at the middle. On the outer surface of the cylinder in a fixed position, detector can also be attached and exposed to radiation. Open ends are covered by perforated sheets. From these three modes the concentrations of radon and thoron gas in Bq m\(^{-3}\) and the potential alpha energy concentration of individual progenies in terms of Working Level Units (WL) are measured.

Three pieces of LR-115 (type II) detectors of size 3 cm x 3 cm were placed in proper positions of the dosimeter cups. The bare detector, mounted on the out side of the cup, views a hemisphere of air of radius at least 9.1 cm, the range of \(^{212}\)Po alpha in air or 6.4 cm, the range of \(^{214}\)Po alpha.\(^7\) It records all the tracks due to radon, thoron and their progenies. In the cup with filter paper mode, the detector was fixed on the dividing wall within the dosimeter cup and the mouth of the chamber on its side was covered with a filter paper. In the other chamber of the cup, the detector was fixed on the other side of the same wall and the mouth of the chamber on this end was covered with a filter paper, a mylar, then a filter paper. Filter paper and mylar do not permit the solid daughter products of thoron to pass through them and partly reduces the rate of diffusion of thoron gas itself due to its short half-life. It has been estimated that 98% of radon penetrates, but thoron does not enter the cup.\(^8,9\)

The preview and the schematic diagram of plastic Twin Chamber dosimeter cup are shown in fig.3.1 and 3.2 below.
Fig. 3.1. Preview of Dosimeter Cup
3.2.3. Selection of Sites and Installation of the Dosimeter Cups:

The following criteria are applied to select the location for placing the dosimeter with detector within a room.

A position is selected where the detector will not be disturbed during the observation period and where there is adequate room for the device. Care is taken not to place the dosimeter in any of the room where there is possibility of air currents. Locations near excessive heat, such as fireplaces or in direct sunlight and areas of high humidity are also avoided.\(^\text{10}\)
The location should not be within 90 centimeters (about 3 feet) of windows or other potential openings in the exterior wall. The dosimeter is hung at an appropriate place within a room of the house in such a way that the dosimeter is not disturbed by inhabitants during the observation period. All dosimeters in the present study are suspended at a height of 2 to 2.5 meters (about 6 to 8 feet) from the floor.

The Plastic Twin-Chamber dosimeter cups with detectors were installed in Reinforced Cement Concrete (R.C.C.) houses in ground floor in such a way that no wall or other surfaces (like roof) is closer than 10 cm. from the detector. The choice of the house was random one and one room in each house was selected for the measurement. Generally sitting rooms, bed rooms of a house are selected for suspending the dosimeters as people spend most of their indoor time in these rooms. The cups were exposed for about 90–95 days after which they were retrieved.

After retrieval, the exposed detectors were taken out from the dosimeter cups and the exposed detectors were immediately kept in a specially designed container. These were chemically etched within 24 hours after retrieval.

3.2.4. Cross Measurements:

As a measure of cross checking the results of measurements, extra dosimeters are placed at the same room in some cases and radon concentrations for both dosimeters compared. In the present study duplicate measurements were made in at least 10% of the total number of locations.
3.2.5. Background Measurements:

Unexposed detector may contain few tracks because of radon impinging on it during its transportation and storage for a long period at a place other than the selected site. Background measurements are necessary for getting accurate track density value at the location where the dosimeter is placed. We measured the background of a statistically significant number of unexposed detectors from each batch for the entire measurement system. Finally the background count is subtracted from the field sample results.

3.3. WORKING FORMULAE:

3.3.1. FOR PLASTIC TWIN-CHAMBER DOSIMETER CUP:

Let $T_1$ and $T_2$ be the track densities registered in membrane and filter mode exposure (as observed through microscope), $d$ is the exposure days, $C_R$ and $C_T$ be the concentrations of radon and thoron in Bqm$^{-3}$, $K_R$ and $K_T$ be the sensitivity factors for radon and thoron gas. Then

$$C_R = \frac{T_1}{dK_R} \quad (3.1)$$

$$C_T = \frac{(T_2 - T_1)}{dK_T} \quad (3.2)$$

Where $K_R = 0.020$ Tcm$^{-2}$d$^{-1}$/Bqm$^{-3}$ and $K_T = 0.019$ Tcm$^{-2}$d$^{-1}$/Bqm$^{-3}$ for radon and thoron gas in the membrane and filter compartments.

The progeny working levels are expressed in WL units. One WL is a measure of the total potential alpha energy exposure of radon/thoron in one litre of air which will result in the ultimate emission by them of $1.3 \times 10^5$ MeV of alpha ray.
energy. ICRP\textsuperscript{12} now defines the WL as $1.300 \times 10^8$ MeV.m\textsuperscript{3}. It also corresponds to $2.08 \times 10^5$ J m\textsuperscript{3} in S.I. units. The progeny working levels can be estimated as

$$WL_R = C_R F_R / 3.7$$  \hspace{1cm} (3.3)

Where

$$F_R = (0.104 F_{R,A} + 0.518 F_{R,B} + 0.37 F_{R,C})$$  \hspace{1cm} (3.4)

and

$$WL_T = C_T F_T / 275$$  \hspace{1cm} (3.5)

Where

$$F_T = 0.908 F_{T,B} + 0.092 F_{T,C}$$  \hspace{1cm} (3.6)

Where $F_R$ and $F_T$ are the equilibrium factors for radon and thoron progeny respectively, corresponding to the extracted ventilation rate.

$$F_{R,A} = D_{R,A} / \{ D_{R,A} + (U_{R,A} \times WLF) + (1 - U_{R,A}) \times WLC + V \}$$  \hspace{1cm} (3.7)

where, $D_{R,A}$ is decay constant of RaA ($= 3.79 \times 10^{-3}$ s\textsuperscript{-1}) and $U_{R,A}$ is the unattached fraction for RaA ($= 0.2$), WLF is the wall loss rate for fine fraction ($= 10$ h\textsuperscript{-1}) and WLC is the wall loss rate for coarse fraction ($= 0.1$ h\textsuperscript{-1}).

$$F_{R,B} = F_{R,B} D_{R,B} / \{ D_{R,B} + (U_{R,B} \times WLF) + (1 - U_{R,B}) \times WLC + V \}$$  \hspace{1cm} (3.8)

Where, $D_{R,B}$ is the decay constant of RaB ($= 4.3 \times 10^{-4}$ h\textsuperscript{-1}) and $U_{R,B}$ is the unattached fraction for RaB ($= 0.025$) and $V$ is the ventilation rate.

$$F_{R,C} = F_{R,B} D_{R,C} / \{ D_{R,C} + (U_{R,C} \times WLF) + (1 - U_{R,C}) \times WLC + V \}$$  \hspace{1cm} (3.9)
Where, \(D_{R-C}\) is the decay constant of RaC (\(^{214}\text{Bi}\)) (= 5.78 \times 10^{-4} \text{s}^{-1}) and \(U_{F_{R-C}}\) is the unattached fraction for RaC (= 0.001).

\[
F_{T-A} = \frac{D_{T-A}}{[D_{T-A} + WLC + V]} \quad (3.10)
\]

Where \(D_{T-A}\) is the decay constant of ThA (\(^{216}\text{Po}\)) (= 1.82 \times 10^{-5} \text{h}^{-1}).

\[
F_{T-C} = \frac{F_{T-A} \times D_{T-C}}{[D_{T-C} + WLC + V]} \quad (3.11)
\]

Where \(D_{T-C}\) is the decay constant for ThC (\(^{212}\text{Bi}\)) (= 1.91 \times 10^{-1} \text{s}^{-1}).

Representative average thoron concentration denoted by \(C_T\),

\[
C_{T-B} = C_T \times F_{T-B} \quad (3.12)
\]

\[
C_{T-B} = C_T \times F_{T-B} \quad (3.13)
\]

Finally, the estimate of the inhalation dose is given by:

\[
D = \frac{(0.17 + 9F_{R})C + (0.11 + 32F_{T})C_T}{1000 \mu \text{Sv.h}^{-1}} \quad (3.14)
\]

A computer program was developed (with the kind help of Dr. T.V. Ramachandran of BARC, Mumbai) in order to carry out these computations. With the help of this computer program we can calculate the radon, thoron and their progeny concentrations, equilibrium factors for radon and thoron, inhalation doses and ventilation rate.

Distribution of indoor radon levels follows a log-normal distribution, which means that there would be a long tail in the distribution where a very small fraction of the total would have very large values. The geometric mean and geometric standard deviation are appropriate for characterising this type of distribution.
3.3.2. For Estimation of Radium Concentrations and Radon Exhalation Rate in Soil Samples:

The “Can technique” is used for the measurement of radium and radon exhalation rates in some soil samples collected from different study areas. The dried samples collected from different places are finely powdered and sieved through a 200 mesh sieve. The fine powder (250g) of samples from each site is placed in different glass bottles and scaled with thin polyethylene sheets for 30 days so as to attain the equilibrium. After one month, LR-115 (type II) plastic track detectors are fixed on the lower side of cork lids, which are then gently pressed against the polyethylene sheets on the glass bottles (acting as emanation chambers) as shown in Fig. 3.4, so that the equilibrium is not disturbed or there is minimum possible disturbance, if any. The bottles are then sealed and left as such for 90 days so that the detectors can record tracks produced by the decay of radon. The exposed detectors are etched in 2.5N, NaOH solution at (60 ±1)°C for 90 minutes as discussed in 3.2.6. The tracks are counted using an (Olympus) optical microscope at 400X magnification.
Fig. 3.3: The Can technique used for the study of radium content and radon exhalation rate of soil samples.

The "Can technique" proposed by\(^{13}\) and later developed by Somogyi\(^{15}\) is used to calculate the radium concentration in soil samples. The radium concentration in soil samples is calculated using the relation,\(^{17}\)

$$C_{RA} = \rho A / KT_e M$$ (3.15)

Where \(C_{RA}\) is the effective radium content of the given sample (Bqkg\(^{-1}\)), \(\rho\) is the track density (track cm\(^{-2}\)), \(M\) is the mass of the sample (250 g), \(A\) is the area of
cross section of bottle \((7.085 \times 10^{-3} \text{ m}^2)\), \(h\) is the distance between the detector and the top of the sample (0.135 m), \(K\) is the sensitivity factor, which is equal to 0.0245 tracks \(\text{cm}^{-2} \cdot \text{d}^{-1} \text{per Bq} \cdot \text{m}^{-3}\) and \(T_e\) is the effective exposure time (in days) which is related with the actual exposure time \(T\) and decay constant \(\lambda\) for \(^{222}\text{Rn}\) with the relation:

\[
T_e = T - \frac{1}{\lambda} \left(1 - e^{-\lambda T}\right) \tag{3.16}
\]

The radon exhalation rate in terms of area is calculated from the equation\(^{19,20}\)

\[
E_A = CV\lambda / A \left[ T + 1 / \lambda (e^{\lambda T} - 1) \right] \tag{3.17}
\]

Where \(E_A\) is the radon exhalation rate in terms of area (Bq.m\(^{-2}\)hr\(^{-1}\)); \(C\) is the integrated radon exposure as measured by LR-115 plastic detector (Bq.m\(^{-3}\)hr); \(V\) is the volume of the can (m\(^3\)); \(\lambda\) \((= 7.5 \times 10^{-3} \text{ hr}^{-1})\) is the decay constant for radon; \(A\) is the area of the can (m\(^2\)). This formula is also modified to calculate the radon exhalation rate in terms of mass (Bq.Kg\(^{-1}\)hr\(^{-1}\)):

The radon exhalation rate in terms of mass is calculated from the expression:

\[
E_m = CV\lambda / M \left[ T + 1 / \lambda (e^{\lambda T} - 1) \right] \tag{3.18}
\]

Where \(E_m\) is the radon exhalation rate in terms of mass and \(M\) is the mass of the sample (250 gm).

The value of \(\lambda\) can be calculated with the help of the formula:

\[
T_{1/2} = 0.693/\lambda \tag{3.19}
\]

Where \(T_{1/2}\) = half life of radon = 3.825 days.
The integrated Radon concentration can be calculated with the help of the formula

\[ C_R = \frac{T_R}{dK} \]  

(3.20)

Where \( T_R \) is the number of tracks \( \text{cm}^{-2}\text{d}^{-1} \), \( d \) is the time of exposure, \( K \) is the calibration factor (Sensitivity factor) = 0.0245 Tracks \( \text{cm}^{-2}\text{d}^{-1}/\text{Bq.m}^{-3} \).

### 3.4.1. Etching Apparatus: (Both for Indoor Radon and Soil Radon Study)

Chemical etching is the important step to make tracks in solid state nuclear track detectors observable by optical microscope. By the method of etching using a suitable chemical etchant, the latent tracks can be enlarged to microscopically visible size. The transformation of a latent track to a visible track is determined by the simultaneous action of two etching processes: a) chemical dissolution along the path of the alpha particle at a faster rate, b) chemical dissolution of the bulk material at a slower rate.

In the present study the exposed films of LR-115 films are etched in 2.5 N NaOH solutions at (60±1) °C for 90 minutes. The solution is kept in a glass beaker of 1000 ml. Now to suspend the detectors in the solution a straw of length 2 cm (Fig.3.4) is taken. In this straw a fine incision is made by a sharp razor blade and in this incision one edge of the detector piece is inserted. This straw is allowed to float in the solution. When the straw floats, the detector inserted in it remains suspended vertically as
Fig. 3.4: Schematic Diagram of Etching Apparatus
Shown in the Fig. 3.4. Normally 6 to 9 detectors are etched at a time in this manner. To ensure uniform etching of the films the solution is subjected to very mild stirring using a magnetic stirrer. The arrangement for etching the detectors is shown in Fig.3.4.

After etching, the films are taken out from the solution and thoroughly cleaned in a jet of distilled water. Then the films are allowed to dry slowly and after that kept in a Desicator. The measurements of optically visible tracks are done by an optical microscope.

3.4.2. Optical microscope:

An optical microscope (Olympus) was used to scan the chemically etched tracks. Various magnifications could be used with different combinations of objectives and eyepieces. Magnification used for the track density measurement in the present study was 400X. The eye piece was fitted with a square graticle which was used to count the tracks randomly all over the detector surface, for 100 fields of view to obtain a representative value. The area of each field of view was 0.004225 cm².

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References:


12. ICRP (1993), Protection against Radon-222 at home and at work. ICRP publication 65, Annals of the ICRP, 23(2).


