5.1 SSNTD-ELECTRET SYSTEM FOR RADON-THORON DOSIMETRY

5.1.1 Introduction

The decay products of Radon and Thoron are known to carry positive charge immediately after their formation (Dua and Kotrappa, 1981). This property has been used by several investigators to collect the decay products on a metal electrode maintained at a high negative potential and assess the concentration of gases by measurement of alpha activity either in situ or subsequently.

It has been recently demonstrated (Kotrappa & co-workers, 1980) that the negatively charged surface of an electret can be used in place of high voltage source for such collection (Electret is an electrical analogue of a permanent magnet, i.e. it is a permanently charged dielectric sheet carrying charges of opposite sign on either side). It produces a constant electrostatic field also. Teflon electrets (6 cm in diameter and 0.08 cm thick) used for collection of decay products produce electrostatic field equivalent to that produced by a battery of 1000 to 3000 volts. In their earlier work, Kotrappa & co-workers (1980) have described a procedure for assessing the concentration of Radon and Thoron from the alpha activity collected on electret when it is exposed in a ten liter wire mesh chamber whose sides are covered with polyurethane foam or Whatman filter paper.

The object of the present work was to adopt the electret
chamber technique for collecting the decay products directly on the surface of a SSITD. This procedure allows registration of alpha activity in situ and hence is useful in determining long term average concentrations.

5.1.2 Electret Chamber

Design of the electret chamber used in these studies is shown in Fig. 5.1. The chamber has a volume of about 10 liters and it is cylindrical in shape. The chamber diameter is twice its height. This minimizes the differences in distances of periphery from the centre of the bottom plate where the electret is located. The chamber top and bottom consist of aluminium plates while the sides are made of wire mesh (20-70 mesh). The wire mesh is covered with polyurethane foam (25 or 75 mm thick) or Whatman filter paper. The wire mesh also helps to avoid the electrostatic interference of the polyurethane foam, if any. The top aluminium cover has an opening through which the electret holder could be introduced or taken out. This opening can be closed with a metal lid. The bottom aluminium plate has a groove in which the brass electret holder sits. The chamber is light weight and portable. It can be left on surface or suspended anywhere by means of hooks provided.

A stainless steel sheet (33 mm diameter and 0.7 mm thick) is fixed on top (centre) of the electret with the help of a small piece of double sided adhesive tape. Thus the metal surface close to the electret's negatively charged surface gets an induced positive charge and a compensating negative charge appears on the top surface of the
FIG 5.1. ELECTRET CHAMBER FOR RADON/THORON MEASUREMENTS.
metal sheet. The charge density on the top surface of the metal sheet is about the same as that on the surface of the electret. The compensating charge (being not bound) is liable to be discharged even by a slight touch with any conducting surface. Therefore, it is necessary to lower the metal sheet with the help of a clean teflon tweezer and further manipulations should be carried out with this tweezer only. The compensating charge stays indefinitely if no ions are deposited on this surface. If the compensating charge leaks away by accidental touching, it is only necessary to remove the metal sheet, ground it and put it back on the surface of the electret using the teflon tweezer.

The above method of using the charge of the electret without directly using the electret has several advantages - (1) the electret remains intact and can be reused, (2) any non-uniform charge distribution on the surface of the electret is not reflected in the charge density distribution on the surface of the metal sheet. This latter advantage is expected to result in more uniform deposition of decay products on the surface of the track detector sheet and hence lead to corresponding improvement in the uniformity of track density distribution.

5.1.3 Detectors

Commercially available sheets of CR-39 plastic (sold under the trade name 'Homalite 6') which are presently known to be the most sensitive SSNTD and also characterised by low background were extensively used in the present work. These detector sheets (1.6 mm thick) were cut into small pieces (size 15 mm x 14 mm) and were covered completely with a thin film (1.0 mg.cm⁻²) of aluminised Mylar with the conducting surface
pointing outwards. This cover attenuates very little of alpha energy but helps in an efficient collection of decay products by not allowing the dielectric properties of the detector material to interfere with the experiment.

An auxiliary experiment was also carried out to determine the alpha track registration efficiency of CR-39 detector sheet (wrapped in the aluminised Mylar of thickness 1.0 mg.cm$^{-2}$) for decay products of Thoron. The decay products of Thoron were collected on the surface of a thin Mylar foil (in an Electret Chamber as described by Kotrappa & co-workers, 1980) and allowed to decay for about ten hours to ensure that subsequent decay followed the half-life of Th-B. A square piece (10 mm x 10 mm) was cut out from this sample and counted carefully, using a calibrated alpha counter, to assess the level of activity on the Mylar film. Two SSNTD pieces covered with aluminised Mylar were put in contact with the remaining piece of Mylar having Thoron decay products and a weight was put on the top of it to ensure intimate contact. The exposure was continued for a period of 24 hours. The expected integrated alpha disintegrations during this interval were calculated from the data on (i) initial alpha disintegration rate, (ii) half-life of Th-B and (iii) the exposure time. Using this procedure and employing the standard track evaluation technique for alpha tracks in CR-39, i.e. etching for 4.5 hours in 6N KOH at 60°C followed by counting of tracks under the microscope, the track registration efficiency of this detector was found to be 40 ± 2.2% (Kotrappa & co-workers, 1981). This data is represented in Table 5.1.
Table 5.1

<table>
<thead>
<tr>
<th>Integral exposure as seen by track detector</th>
<th>No. of tracks counted and the no. of views scanned*</th>
<th>Corresponding track density in no. cm⁻²</th>
<th>Track registration efficiency</th>
</tr>
</thead>
<tbody>
<tr>
<td>61683 per cm² tracks in 143 views</td>
<td>1988 ± 44</td>
<td>24648 ± 2.2 %</td>
<td>40 ± 2.2 %</td>
</tr>
</tbody>
</table>

* Sum of the numbers for two detector films

5.1.4 Experiments and Results

For controlled generation of Radon and Thoron the arrangement shown in Fig. 5.2 is employed. Air passing through a chamber containing a leaky Radium needle is mixed with humidity conditioned air (HCA) and injected into an inverted half cut carbouy. The top portion of the cut carbouy is covered with a 25 mm thick polyurethane foam. Air containing Radon enters the drum through this layer of foam and finally escapes out through a perforated cover on the drum. Electret chamber is suspended in the drum at the centre. Humidity and temperature can be measured continuously in the drum. For experiments with Thoron, a chamber containing 6-10 g of Thorium hydroxide packed in double layer of filter is used in place of the chamber containing leaky Radium needle. The concentration of Radon/Thoron can be varied by regulating the flow of the conditioned air.
FIG. 5.2. ARRANGEMENT FOR CONTROLLED GENERATION OF RADON/THORON ATMOSPHERES
To carry out the experiments, the electret chamber was first hung inside the drum and then the generation of Radon/Thoron started. After allowing a few minutes for the attainment of equilibrium/steady-state conditions inside the chamber, the electret holder along with SSNIIID piece was lowered in the chamber and its top closed with the metal lid. A small 'double' filter unit (IAEA, 1976) was also introduced in the drum with its sampling point mid-way between the top and the bottom of the electret chamber, for the measurement of the Radon/Thoron concentration. It sampled the air at a flow rate of 5 liter/min continuously throughout the period of exposure of the detector. The total flow of air through the drum was 70 liter/min. The relative humidity and the temperature during the experiments were 30% and 25°C respectively.

At the end of the sampling period, the electret was removed and the status of charge on the electret checked. The final charge should not be below $10^{-9}$ coulomb/cm$^2$ for satisfactory performance (The initial charge is usually in the range of $5-10 \times 10^{-9}$ coulomb/cm$^2$). The detector samples were allowed a delay of at least 3 hours and 3 days before etching in the case of Radon and Thoron respectively. This delay allows the registration of all the possible alpha emissions from the decay products collected on the surface of SSNIIID and makes the calibration factor independent of time of sampling/exposure (this will be clear from the results).

The double filter sample was analysed by the standard procedure to calculate the Radon or Thoron concentration. This
concentration is given by the equation:

\[ C = \frac{0.45 X}{E \cdot F \cdot Z \cdot V} \]

where

- \( C \) = concentration of Radon/Thoron in pCi.lit\(^{-1}\).
- \( X \) = integral alpha counts obtained between the time intervals \( t_1 \) and \( t_2 \) reckoned from the end of sampling.
- \( E \) = counting efficiency of the alpha counter.
- \( Z \) = theoretical buildup and decay term for the decay products of Radon or Thoron and is a function of \( t_1, t_2 \) and \( t_s \) (\( t_s \) = duration of sampling). These values are available in literature (Thomas, 1970, 1971).
- \( V \) = volume of the double filter chamber in liters.
- \( F_e \) = fraction of the decay products collected on the 2nd filter paper, of the total produced inside the decay chamber. This is also known as collection efficiency.

Table 5.2 and Table 5.3 give the results on the response of the track detectors to Radon and Thoron respectively. It is seen that for a given cumulative concentration \( \int \text{(pCi.lit}^{-1}) \cdot h \), the response is independent of the actual concentration or the time of exposure. This follows from the procedure which allows for the complete decay of the daughter products accumulated on the surface of SSNTD.
Table 5.2
Response of SSKTD (CR-39) to Radon in Electret Chamber

<table>
<thead>
<tr>
<th>Expt. no.</th>
<th>Conc. of Radon in ( \text{Ci.lit}^{-1} )</th>
<th>Time of sampling</th>
<th>No. of tracks counted</th>
<th>Track density ( \text{no/cm}^2 )</th>
<th>( % )</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>190</td>
<td>1.0</td>
<td>512</td>
<td>14436</td>
<td>76 ± 4.4%</td>
</tr>
<tr>
<td>2</td>
<td>191</td>
<td>1.0</td>
<td>363</td>
<td>16745</td>
<td>96.1 ± 5.2%</td>
</tr>
<tr>
<td>3</td>
<td>192</td>
<td>1.0</td>
<td>938</td>
<td>16756</td>
<td>87.3 ± 3.3%</td>
</tr>
<tr>
<td>4</td>
<td>116</td>
<td>0.5</td>
<td>544</td>
<td>12364</td>
<td>106.6 ± 4.3%</td>
</tr>
<tr>
<td>5</td>
<td>317</td>
<td>1.0</td>
<td>1637</td>
<td>28240</td>
<td>89.1 ± 2.5%</td>
</tr>
<tr>
<td>6</td>
<td>642</td>
<td>2.0</td>
<td>4161</td>
<td>54902</td>
<td>85.5 ± 1.6%</td>
</tr>
<tr>
<td>7</td>
<td>798</td>
<td>3.0</td>
<td>1633</td>
<td>68926</td>
<td>86.4 ± 2.5%</td>
</tr>
<tr>
<td>8</td>
<td>185</td>
<td>0.75</td>
<td>512</td>
<td>14879</td>
<td>80.4 ± 4.4%</td>
</tr>
<tr>
<td>9</td>
<td>124</td>
<td>0.77</td>
<td>535</td>
<td>13745</td>
<td>110.8 ± 4.3%</td>
</tr>
<tr>
<td>10</td>
<td>154</td>
<td>0.75</td>
<td>537</td>
<td>10577</td>
<td>68.7 ± 4.3%</td>
</tr>
<tr>
<td>11</td>
<td>136</td>
<td>0.75</td>
<td>567</td>
<td>13226</td>
<td>97.2 ± 4.2%</td>
</tr>
<tr>
<td>12</td>
<td>113</td>
<td>0.75</td>
<td>572</td>
<td>10242</td>
<td>90.6 ± 4.2%</td>
</tr>
<tr>
<td>13</td>
<td>143</td>
<td>0.75</td>
<td>578</td>
<td>11513</td>
<td>80.5 ± 4.2%</td>
</tr>
<tr>
<td>14</td>
<td>127</td>
<td>0.75</td>
<td>588</td>
<td>13364</td>
<td>105.2 ± 4.1%</td>
</tr>
<tr>
<td>15</td>
<td>222</td>
<td>1.13</td>
<td>462</td>
<td>17063</td>
<td>76.9 ± 4.6%</td>
</tr>
<tr>
<td>16</td>
<td>157</td>
<td>0.83</td>
<td>540</td>
<td>16225</td>
<td>103.3 ± 4.3%</td>
</tr>
<tr>
<td>17</td>
<td>159</td>
<td>1.0</td>
<td>510</td>
<td>17058</td>
<td>107.3 ± 4.4%</td>
</tr>
<tr>
<td>18</td>
<td>157</td>
<td>1.07</td>
<td>528</td>
<td>17660</td>
<td>112.5 ± 4.4%</td>
</tr>
</tbody>
</table>

Grand mean and standard deviation of \( \% \):

\[ \bar{\%} = 92.4 ± 13.3 \%
\]
\[ \% = 92.4 ± 14.3 \%
\]

Note:
(1) The track detectors were etched after a minimum delay of 3 hours after sampling. Relative humidity and temperature were 30% and 25°C respectively.

(2) To express concentration in now units, use the following conversion factor: 1 pCi.lit \( ^{-1} \) = 37 Bq.m \( ^{-3} \)
Table 5.3
Response of SSNTD (CR-39) to Thoron in Electret Chamber

<table>
<thead>
<tr>
<th>Expt. no.</th>
<th>Conc. of Thoron in (pCi.lit^{-1} . h)</th>
<th>No. of tracks counted</th>
<th>Track density (no. cm^{-2})</th>
<th>T/C</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>950</td>
<td>513</td>
<td>9579</td>
<td>10.3 ± 4.4 %</td>
</tr>
<tr>
<td>2</td>
<td>1100</td>
<td>560</td>
<td>13783</td>
<td>12.5 ± 4.2 %</td>
</tr>
<tr>
<td>3</td>
<td>1188</td>
<td>602</td>
<td>11120</td>
<td>9.4 ± 4.1 %</td>
</tr>
<tr>
<td>4</td>
<td>1100</td>
<td>511</td>
<td>10193</td>
<td>8.6 ± 4.4 %</td>
</tr>
<tr>
<td>5</td>
<td>1010</td>
<td>515</td>
<td>7737</td>
<td>7.7 ± 4.4 %</td>
</tr>
<tr>
<td>6</td>
<td>1342</td>
<td>599</td>
<td>13796</td>
<td>10.3 ± 4.1 %</td>
</tr>
<tr>
<td>7</td>
<td>1183</td>
<td>564</td>
<td>9523</td>
<td>8 ± 4.2 %</td>
</tr>
<tr>
<td>8</td>
<td>1142</td>
<td>523</td>
<td>10906</td>
<td>9.6 ± 4.4 %</td>
</tr>
<tr>
<td>9</td>
<td>1174</td>
<td>517</td>
<td>11047</td>
<td>9.4 ± 4.4 %</td>
</tr>
<tr>
<td>10</td>
<td>1376</td>
<td>526</td>
<td>10513</td>
<td>7.6 ± 4.4 %</td>
</tr>
<tr>
<td>11</td>
<td>1329</td>
<td>509</td>
<td>9396</td>
<td>7.1 ± 4.4 %</td>
</tr>
<tr>
<td>12</td>
<td>864</td>
<td>613</td>
<td>7102</td>
<td>8.2 ± 4 %</td>
</tr>
</tbody>
</table>

Grand mean and standard deviation of T/C = 9.1 ± 1.5
= 9.1 ± 16.7 %

Note: (1) Track detectors were etched after a minimum delay of 3 days after sampling. Relative humidity and temperature were 30% and 25°C respectively. Sampling duration was one hour in all the cases.

(2) To express concentration in new units, use the following conversion factor: 1 pCi.lit^{-1} = 37 Bq.m^{-3}. 
5.1.5 Discussion

From Table 5.2, it is seen that on an average, one can expect 92 tracks cm$^{-2}$ per $(\text{pCi} \cdot \text{lit}^{-1}) \cdot h$ for Radon. Taking 500 tracks cm$^{-2}$ as the lower limit of track density which can be conveniently evaluated using a microscope, the minimum measurable concentration of Radon works out to about $0.23 \text{ pCi} \cdot \text{lit}^{-1}$ for a sampling period of 24 hours. Still lower levels can be estimated by increasing the duration of sampling.

The sensitivity of Thoron is seen from Table 5.3 to be about $9.4 \text{ tracks cm}^{-2}$ per $(\text{pCi} \cdot \text{lit}^{-1}) \cdot h$. This is very much lower than that for Radon for the following reason: The concentration of Thoron inside the electret chamber is considerably lower than that outside where the double filter is located. This is due to short half-life of Thoron.

The above measurements for Thoron were carried out using Whatman filter paper to cover the wire mesh screen of the electret chamber. It was observed that if 75 mm thick foam was used instead, it led to complete decay of Thoron before it reached the sensitive volume of the electret chamber. This foam thickness had, however, no effect on the Radon sensitivity except that it took a little longer time for the attainment of equilibrium/steady state conditions. This effect can, therefore, be conveniently exploited for the simultaneous measurement of Radon and Thoron by using two chambers side by side, one covered with Whatman filter paper and the other covered with 75 mm thick foam.

It must also be borne in mind that the sensitivity of this method depends to some extent on the relative humidity (Dua & co-workers,
1983) of the sampling environment. It is however possible to apply
correction for the same as has been reported earlier (Kotrappa &
co-workers, 1980). Electrets also collect ions produced by ionisation
due to external radiation causing lowering of strength of the electric
field. However, this does not pose any problem at the normal background
levels of 50 to 100 μR.h⁻¹ for sampling periods upto three days. If
the sampling has to be done for a longer period, it is only necessary
to remove the metal sheet, ground it and put it back onto the electret
using a teflon tweezer. Same detectors can be placed on the metal
sheet again to continue sampling.

The possibility of using CR-39 SSNTD for recording alphas
directly from an atmosphere containing Radon has been reported earlier
(Cassou and Benton, 1978). This approach has been tried by others and
a sensitivity of 0.1264 tracks.cm⁻² per (pCi.lit⁻¹).h⁻¹ for Radon has
been reported (Fleischer & co-workers, 1980). In this method there is
no collection of decay products on the SSNTD surface. The sensitivity
which arises from the contact of SSNTD with ambient air, is therefore
low. This is the consequence of the sensitive volume, from which alphas
can register their tracks, being quite small. When we compare our
sensitivity value for Radon with the one just mentioned, a sensitivity
improvement by a factor of about 730 is achieved. Similar comparison
could not be made for Thoron as corresponding figures were not
available in literature. This may be because of the fact that
measurements for Thoron have not attracted enough attention or else the
sensitivity value for Thoron may be too small to be measured accurately
by conventional methods.
The sensitivity improvements for Kidon and Thoron were possible merely because of the use of electrets. The above measurements were carried out at levels which are high compared to those prevailing in the environment. Effort is now being made to confirm the utility of the method in actual field conditions.

5.1.6 Limits of Detection

assuming that the above method is suitable for measurement of environmental concentrations of Kidon and Thoron, the minimum level of detection will be governed by the background track densities encountered in various track detector sheets.

If $C_m$ denotes the minimum detection limit of concentration of Kidon or Thoron in units of $(\text{pCi.lit}^{-1}) \cdot \text{h}$ for which the fractional error $E = \frac{r}{C}$ is 0.5, i.e. 50%, then $C_m$ is given by (Thomas, 1971, see Appendix, Ch.5),

$$C_m = \frac{2 (1 + \sqrt{1 + 2B})}{K}$$

where $B = \text{Background track density in tracks} \cdot \text{cm}^{-2} \text{ in the etched detector sheet}$,

and $K = \text{Constant of proportionality} = \frac{92.4 \text{ tracks} \cdot \text{cm}^{-2}}{(\text{pCi.lit}^{-1}) \cdot \text{h}}$ in our case;

knowing the values of $B$ and $K$, $C_m$ can be determined for any detector sheet.

A number of CR-39 track detector sheets obtained from various manufacturers were etched under standard conditions and examined under
the microscope to determine their respective minimum detection limits. This limit (for each side) is given in Table 5.4.

Table 5.4
Minimum detection limits of various CR-39 track detector sheets

<table>
<thead>
<tr>
<th>Sl. No.</th>
<th>Source of detector and specifications if any</th>
<th>Side examined</th>
<th>No. of tracks counted/No. of views scanned</th>
<th>Background track density no. cm^-2 (= B) &amp; the standard deviation</th>
<th>Minimum limit &quot;c&quot; in mCi.lit. h^-1</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Pershore Mouldings, U.K. 16 h curing</td>
<td>i)</td>
<td>too much background</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>ii)</td>
<td>11/404</td>
<td>48 ± 30.2 %</td>
<td>0.24</td>
</tr>
<tr>
<td>2</td>
<td>Pershore Mouldings, U.K. 32 h curing</td>
<td>i)</td>
<td>6/518</td>
<td>21 ± 40.8 %</td>
<td>0.16</td>
</tr>
<tr>
<td></td>
<td></td>
<td>ii)</td>
<td>12/468</td>
<td>45 ± 28.9 %</td>
<td>0.23</td>
</tr>
<tr>
<td>3</td>
<td>Pershore Mouldings, U.K. 96 h curing</td>
<td>i)</td>
<td>48/425</td>
<td>200 ± 14.4 %</td>
<td>0.46</td>
</tr>
<tr>
<td></td>
<td></td>
<td>ii)</td>
<td>35/509</td>
<td>122 ± 16.9 %</td>
<td>0.36</td>
</tr>
<tr>
<td>4</td>
<td>American Acrylics, U.S.A. (received from a scientist colleague)</td>
<td>i)</td>
<td>43/341</td>
<td>223 ± 15.2 %</td>
<td>0.48</td>
</tr>
<tr>
<td></td>
<td></td>
<td>ii)</td>
<td>78/451</td>
<td>306 ± 11.3 %</td>
<td>0.56</td>
</tr>
<tr>
<td>5</td>
<td>American Acrylics, U.S.A.</td>
<td>i)</td>
<td>44/495</td>
<td>157 ± 15.1 %</td>
<td>0.41</td>
</tr>
<tr>
<td></td>
<td></td>
<td>ii)</td>
<td>21/378</td>
<td>98 ± 21.8 %</td>
<td>0.33</td>
</tr>
<tr>
<td>6</td>
<td>E3G Industries, U.S.A.</td>
<td>i)</td>
<td>too much background</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>ii)</td>
<td>9/462</td>
<td>35 ± 33.3 %</td>
<td>0.2</td>
</tr>
<tr>
<td>7</td>
<td>*Homalite, U.S.A.</td>
<td>i)</td>
<td>51/400</td>
<td>226 ± 14 %</td>
<td>0.48</td>
</tr>
<tr>
<td></td>
<td></td>
<td>ii)</td>
<td>not examined</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

* In some of the pieces of this detector, which were stored in paper envelopes for more than a year, the background observed was 600-700 tracks cm^-2.
From this data, it is clear that detector sheets are available which can measure Radon concentrations as low as 0.01 pCi.lit$^{-1}$ for sampling periods of 24 hours, i.e., it should be possible to measure conveniently the environmental Radon concentrations. Similarly, it should be possible to measure Thoron concentrations which are about ten times higher (because K for Thoron is equal to $\frac{9.1 \text{ tracks cm}^{-2}}{(\text{pCi.lit}^{-1}).\text{h}}$).

Having confirmed the usefulness of various detector sheets, field experiments are now being planned and the results will be reported in due course.

5.2 GAMMA DOSIMETRY IN THE MEGA-RAD (Kilo-Gray) REGION

5.2.1 Introduction

Gamma dosimetry in the mega-rad (kGy) region is also included in the various fields, where the track recording plastics have played an important role. The method is based on the fact that gamma radiation produces damage in the bulk of these plastics which can be measured by different physical or chemical means. The importance of these plastics arises from the fact that they satisfy the basic dosimetric requirements (Pardue & co-workers, 1944; Davis & co-workers, 1954) of low cost, simplicity, and high degree of reproducibility.

Day and Stein (1951) suggested the use of changes in optical density of plastics, due to irradiation (or absorbed dose) as a means of dosimetry. Such changes have been observed for polycarbonate films (Boag & co-workers, 1958) due to gamma irradiation. Benton (1968)
suggested the dependence of bulk chemical etch rate on absorbed dose rate and has shown that Gn plastic can be used to measure doses in the range of $10^5 - 10^6$ rads. Similarly Lexan polycarbonate plastic has also been investigated (Frank and Benton, 1970) and found useful in the range of $10^7 - 10^9$ rads. The utility of Daicoll (trade mark of Daicel Ltd., Japan for GN film) has also been established in the range of $10^5 - 10^7$ rads (Ram and Bose, 1982).

In the work reported here, Makrofol N (195 μm thick) was studied for gamma dosimetry using both optical density and bulk chemical etch rate methods.

The plastic pieces ($1" \times 1"$ squares or $1"$ diameter circles) were successively washed with detergent, tap water and distilled water, dried with filter paper and handled subsequently with tweezers only. Irradiation of plastic pieces was carried out in a cobalt-60 gamma chamber having a dose rate of $1.2 \times 10^5$ rad h$^{-1}$.

5.2.2 Dosimetry by Optical Density (O.D.) Method

O.D. measurements were carried out using Cary recording spectrophotometer, model-14 which has an O.D. range of 0.01 to 2.0. The plastic pieces ($1" \times 1"$) can be easily accommodated in both the sample and the control compartments.

When irradiated pieces of Makrofol were examined in the visible and UV region, against air in the control compartment, no useful information could be obtained. This was because there was practically no difference between unirradiated and irradiated pieces in the visible region, while in the UV region the unirradiated pieces themselves showed
tremendous absorption so that any change in absorption due to irradiation was not easily noticeable.

To overcome this difficulty, irradiated pieces were studied against unirradiated pieces as controls. This brought out the effect of irradiation clearly as shown in Fig. 5.3. A clear peak is found to exist at about 284 nm. This peak O.D. value was found to increase with absorbed dose. All subsequent measurements were therefore carried out in the region corresponding to this peak. This not only gives the maximum value of O.D. for the given absorbed dose but also minimises the effect of errors due to wavelength shift or slit width. The enhanced UV absorption is a direct consequence of the new chemical species formed as a result of energy deposition by the incident gamma-ray. These species differ from the bulk polymer in the sense that they possess electronic configurations which strongly absorb UV radiation at that wavelength (Benton, 1970).

The O.D. was measured for each piece before and after irradiation (against a standard control piece). The dose range covered was $0.6 \times 10^6$ rads (6 kGy) to $10.8 \times 10^6$ rads (108 kGy) and the $^{60}$Co gamma chamber mentioned above was used for this purpose. When the maximum net O.D. value for each irradiation was plotted against the corresponding absorbed dose, a linear relationship was found to exist which could be expressed by the equation:

$$Y = 0.1714X$$

where $Y =$ O.D. of the piece due to irradiation, and $X =$ absorbed dose in mega-rads.
FIG. 5.3. RADIATION INDUCED ULTRAVIOLET ABSORPTION IN MAKROFOL N (195 µm)
The above equation can be used to measure absorbed doses or dose rates for sources of unknown strengths. No fading in the signal was noticed for periods longer than two months. It is also possible to extend the range of measurements on both higher and lower side by using thinner and thicker films respectively. By using 40 μm thick film doses as high as \(40 \times 10^6\) rads (400 kGy) could also be measured (Shah & co-workers, 1978).

5.2.3 Dosimetry by Etch Rate Method

In these experiments plastic circles of 1.0" diameter were used. They were then irradiated in the manner mentioned above in the dose range of \(10 \times 10^6\) rads (100 kGy) to \(250 \times 10^6\) rads (2500 kGy). Towards the upper end of this dose range, the plastic pieces were found to become sticky. Aluminium foils had to be used, therefore, as spacers during irradiation. Beyond this range the plastic pieces became brittle and difficult to handle. The studies, therefore, could not be extended to higher doses.

All the plastic pieces were weighed before and after irradiation (but before etching) to check if there was any weight loss due to irradiation. It was observed that at doses above \(50 \times 10^6\) rads (500 kGy), there were measurable weight losses which are assumed to be due to escaping gases (Frank and Benton, 1970) produced as a result of irradiation.

Chemical etching was carried out at 80°C in 6N NaOH. The standard etching assembly described in Chapter 2 earlier was used for this purpose. During etching, each circle was held in a small separate compartment formed by interconnecting perspex rings and stainless steel
wire mesh. The weight losses for etching periods of 1, 2, 3 and 3.5 hours were measured. From these values, thickness losses from a single surface were also calculated for each dose. When the average thickness losses for four films were plotted against various etching periods for each dose, straight lines were obtained in all the cases. This data is shown in Fig. 5.4. This indicates that the etch rate remains constant at various etching periods, i.e. the etch rate does not change with depth in the film.

Similarly, when the average thickness losses from a single surface were plotted against the corresponding absorbed doses on a semilog paper for each etching period, parallel straight lines were obtained. These are shown in Fig. 5.5. This indicates that average thickness loss of an irradiated film is an exponential function of the absorbed dose. These parallel straight lines (on semilog paper) can be represented by the general equation (Shah & co-workers, 1978):

\[ Y = 0.00246 X + C \]

where

- \( Y \) = average thickness lost in \( \mu \)m from a single surface (on log scale)
- \( X \) = absorbed dose in mega rads (on linear scale), and
- \( C \) = a constant having values of 0.524, 0.608, 0.960 and 1.028 for etching periods of 1, 2, 3 and 3.5 hours.

5.2.4 Conclusion

From the above studies, it is clear that Makrofol N is a useful dosimeter for the dosimetry of gamma radiation in the mega-rad (kGy) region. The dose ranges encompassed by 195 \( \mu \)m thick film in the
ETCHING IN 6N NaOH AT 80°C

<table>
<thead>
<tr>
<th>CURVE No.</th>
<th>DOSE</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.0 M RAD</td>
</tr>
<tr>
<td>2</td>
<td>33.9</td>
</tr>
<tr>
<td>3</td>
<td>61.5</td>
</tr>
<tr>
<td>4</td>
<td>91.8</td>
</tr>
<tr>
<td>5</td>
<td>119.0</td>
</tr>
<tr>
<td>6</td>
<td>153.2</td>
</tr>
<tr>
<td>7</td>
<td>183.1</td>
</tr>
<tr>
<td>8</td>
<td>214.0</td>
</tr>
<tr>
<td>9</td>
<td>250.0</td>
</tr>
</tbody>
</table>

FIG. 5.4 VARIATION OF THICKNESS LOST WITH ETCHING TIME FOR VARIOUS DOSES FOR MAKROFOL N
FIG. 5.5 DEPENDENCE OF THICKNESS LOST (ETCHRATE) ON THE ABSORBED DOSE
two methods are 1-10 and 10-250 mega-rads (10-100 and 100-2500 kGy) respectively, i.e., these methods supplement each other and enhance the range over which the dosimeter can be employed.

5.3 TRACK ETCHED MICROFILTERS

5.3.1 Introduction

The technique which led to development of track etched microfilters, was discovered entirely accidentally in 1962 by Price and Walker. They observed fine holes due to fission fragments in 12 μm thick layers of synthetic mica during the course of their studies on chemical etching of charged particle tracks. The existence of these holes was demonstrated by using transmission electron microscopy. The chemical etching thus permits drilling of fine holes of adjustable size in thin sheets of many cleavable solids and a number of plastics, which are obtainable commercially in thin sheet form.

These microfilters have uniform holes and thus offer certain advantages over conventional filters made of cellulosic plastics and having irregular holes (Sabin, 1973). Figure 5.6 is a microphotograph of (a) millipore filter paper and (b) microfilter. Because of their distinct advantages, their use in various filtration and separation processes is increasing rapidly.

5.3.2 Formation of Track Holes

The heavy charged particles, passing through any of the solids comprising SSNTD, produce continuous damage along their path and thus leave behind a trail of radiation damaged material. The chemical
FIG. 5.6. MICROPHOTOGRAPH OF (a) MILLIPORE FILTER AND (b) MICROFILTER.
etching of these irradiated solids leads to formation of fine hollow channels along the path of the charged particles due to preferential etching of the damage trail while the rest of the material is essentially unaltered. These channels are usually uniform in width along their entire length and maintain the directions of original tracks (Fleischer & co-workers, 1963). If, therefore, the thickness of the detector is less than the charged particle's range in it, the above process leads to formation of fine holes in the irradiated detector sheet.

5.3.3 Alignment of Holes

In order to obtain a good filter, it is necessary to have all the holes as identical as possible as regards shape, size, length, etc. If the irradiation is such that charged particles (fission fragments in our case) enter the film at all angles, the resulting holes after etching will have little similarity amongst themselves. To avoid this, it was necessary to align the holes and thus improve the quality of the filter. The holes could be aligned by employing a collimated beam of charged particles. A simple way (Fleischer & co-workers, 1963) of collimating a beam of particles was to separate the source from the target film and evacuate the intervening space to overcome degradation in energy of charged particles. The larger the separation and smaller the source area, the more complete is the source alignment.

5.3.4 Experimental Irradiation Assembly

Figure 5.7 gives the simple device used by us for carrying out irradiations. It consists of a vacuum dessoricator which can conveniently
FIG. 5.7. IRRADIATION ASSEMBLY FOR THE PREPARATION OF MICROFILTERS.
hold a vacuum of the order of 0.1 Torr (≈ 100 μm). An electrodeposited $^{252}$Cf source (10 mm diameter) is used as a source of fission fragments. For our irradiations, the distance between the source and the filter was fixed as 12 cm. Since the diameter of the irradiated circular portions is 2.5 cm, the angle of incidence of fission fragments is automatically restricted to within 12° of the normal to the filter sheet. Makrofol sheets of 10, 12 and 16 μm thickness were employed in these investigations. All the etchings were carried out at 60°C in 6N NaOH (Bhagwat and Soman, 1979). A one-to-one correspondence has been established between fission fragment tracks and etched holes under conditions approximating to normal incidence.

5.3.5 Hole Size as a Function of Etching Time

It may be remembered that through holes start developing only after an initial latent period of etching during which etching is taking place, but through holes have not resulted. Once a hole is formed the increase in its diameter is very rapid in the beginning corresponding to the dissolution of the damaged region, after which the increase in diameter follows the bulk etch rate of the undamaged solid. These points will be clear from Fig. 4.26 which represents the variation of size of holes (diameters) with etching time (each diameter value represents an average of 40 measurements). The extrapolated region of the curve is shown by dotted line and is difficult to establish experimentally unless one employs sensitive physical methods of measurement, e.g. electrical resistivity of pores (Bean and Desorbo, 1975).

The largest hole size that can be obtained depends on film
thickness. Thus holes of 9.6, 10.6 and 14.2 μm diameters were obtained in films of 10, 12 and 16 μm thickness. In these large hole size regions, the films become very thin and pose handling problems. With the aid of optical microscopes, we were able to measure conveniently hole diameters as small as 1 μm.

5.3.6 Limitation on Hole Density

Although the upper limit of hole densities obtainable depends on particle fluence, randomness in positions of holes must be considered as a limiting factor. This leads to formation of large sized holes due to overlap. The overlap will increase with increasing porosity and impair the efficiency of the filter in separating particles of given size.

Assuming randomness in positions of holes (with equal chances for any position) and considering the probabilities of overlap in the formation of successive holes, i.e. first, second, third, .... r^{th} .... n^{th}, (n + 1)^{st} hole in a unit area, it is observed that the probability 'p' of overlap in the formation of (n+1)^{st} hole is given by (Bhagwat & co-workers, 1976a):

\[ p = \sum_{r=1}^{n} (-1)^{r+1} \cdot \binom{n}{r} \cdot a^r \quad \ldots \ldots \ldots (1) \]

where \( a \) = average area of a hole in the microfilter. Probability 'p' of overlap here is the same as probability 'p' of loss of a hole during spark counting as described in Chapter 3.
\[ (1-p) = 1 - \sum_{r=1}^{n} (-1)^{r+1} \cdot \binom{n}{r} a^r \]

\[ = (1-a)^n \]

or \[ n = \log \frac{1-p}{\log (1-a)} \]

If \( n \) is taken to represent the hole density, then from equation (2) it is clear that the maximum hole density obtainable is governed by both, the acceptable probability of overlap and the hole size. The Fig. 5.8 gives the various percentage probabilities of overlap when both these factors are considered. From this figure one can choose the filter specifications and make them accordingly.

From equations (1) and (2) given above, it is also seen that

\[ p = 1-(1-a)^n \]

\[ = \text{area occupied by 'n' holes} \]

i.e. probability 'p' of overlap in the formation of \((n+1)\)th hole is equal to the area occupied by 'n' holes. Hence the area occupied by \((n+1)\) holes is just slightly greater than the probability 'p' of overlap in the formation of \((n+1)\)th hole irrespective of hole size.

In any work, involving the use of these microfilters, it is extremely unlikely that filters with overlap probabilities exceeding 0.1 (= 10%) shall be used. The strength of the filter sheet will not be affected significantly by porosity corresponding to this probability and hence need not cause any concern.

5.3.7 Applications

The microfilters, prepared from 10 \(\mu\)m thick Makrofol sheet
FIG. 5.8. GRAPH SHOWING THE PERCENTAGE PROBABILITIES OF OVERLAP OF HOLES IN THE FILTER AS A FUNCTION OF HOLE DIAMETER AND HOLE DENSITY.
in the manner described above, were tested and found useful in the following applications (Bhagwat and Soman, 1979).

(A) **Biomedical Applications**

i) Separation of lymphocytes from tumor cell suspension prepared from solid tumor.

ii) Separation of bacteria from tumor cell suspension.

Fig. 5.9(a) is a microphotograph of Murine Fibrosarcoma Tumor cells (Bhagwat and Soman, 1979) separated by the use of microfilter with hole diameter = $2.86 \mu m$ and hole density = $3.5 \times 10^5 \text{ cm}^{-2}$.

(B) **Environmental Studies of Particulates**

With about 10% porosity, these microfilters are capable of sampling upto 40 litres of air per minute ($= 14 \text{ liters.min}^{-1} \cdot \text{cm}^{-2}$ area of the microfilter) using Rotovac TR-7 type of air sampling pumps and should be of great value in the sampling of atmospheric aerosols.

Fig. 5.9(b) is a microphotograph of atmospheric dust particulates (Bhagwat and Soman, 1979) collected on the microfilter with hole size $= 4.6 \mu m$ and hole density $7.2 \times 10^5 \text{ cm}^{-2}$.

Several applications of these filters have been reported in literature (Fleischer & co-workers, 1975), e.g. in cleaning of gases, to clarify wine and beer, in filtration of drugs, in immunology and in the study of phenomenon of superfluidity and superconductivity, to mention a few. It is intended to carry out further studies of additional applications in our laboratory.

5.3.8 **Micro-Filters from Other Materials**

We were also successful in preparing microfilters from thin sheets of mica and locally made CN films. Holes in mica were
MICROPHOTOGRAPHS OF (a) MURINE FIBROSARCOMA TUMOR CELLS SEPARATED BY THE USE OF MICROFILTER (HOLE \( \phi = 2.9 \mu m \)) (b) ATMOSPHERIC DUST PARTICULATES COLLECTED ON THE MICROFILTER (HOLE \( \phi = 4.6 \mu m \))

FIG. 5.9.
rhombi in shape with angles of 60° and 120°. Here the hole sizes are mentioned by giving diameters of equivalent area circles. These filters should prove useful in work at elevated temperatures, because mica withstands high temperatures easily. An application of this type has been reported recently (Tress & co-workers, 1981).

For CN films it was also possible to carry out irradiation with alpha particles of energy 2-3 MeV. The only limitation with filters made from CN films is that they are not as strong as those made from Mikrofol.

5.3.9 Recent Improvements

When fission fragments are used for irradiations, the resulting filters show a small distribution in hole sizes. This is because fission fragments have a range of distribution in mass and energy. To overcome this shortcoming irradiations are now being carried out using accelerator produced monoenergetic heavy ion beams (Fischer and Spohr, 1980). The resulting filters have holes of the same size.

5.4 AUTORADIOGRAPHY OF UO₂ - PuO₂ FUEL PELLETS

5.4.1 Introduction

A programme of preparation of mixed oxide fuels was initiated, some time back, at BARC to evaluate their utility for reactor applications. The initial lots had a composition of 97.4 % UO₂ and 2.6 % PuO₂ by weight. The physical characteristics of these fuel pellets are given in Table 5.5.
Table 5.5

Physical characteristics of fuel pellets

<table>
<thead>
<tr>
<th>i)</th>
<th>Diameter</th>
<th>14.5 mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>ii)</td>
<td>Height</td>
<td>21 mm</td>
</tr>
<tr>
<td>iii)</td>
<td>Weight</td>
<td>29.6 g</td>
</tr>
<tr>
<td>iv)</td>
<td>Density</td>
<td>10.56 g/cm³ (= 96 % of the theoretical value)</td>
</tr>
<tr>
<td>v)</td>
<td>Average size of the individual particles</td>
<td>2-5/μm</td>
</tr>
<tr>
<td>vi)</td>
<td>Composition of Plutonium</td>
<td></td>
</tr>
<tr>
<td>$^{239}\text{Pu}$</td>
<td>94.25 % (by wt.)</td>
<td></td>
</tr>
<tr>
<td>$^{240}\text{Pu}$</td>
<td>5.35 % &quot;</td>
<td></td>
</tr>
<tr>
<td>$^{241}\text{Pu}$</td>
<td>0.339 % &quot;</td>
<td></td>
</tr>
<tr>
<td>$^{242}\text{Pu}$</td>
<td>0.016 % &quot;</td>
<td></td>
</tr>
<tr>
<td>$^{238}\text{Pu}$</td>
<td>0.014 % &quot;</td>
<td></td>
</tr>
</tbody>
</table>

Although the two oxides had been thoroughly mixed, it was preferable to confirm the same by some means. It was with this aim that the autoradiography of fuel pellets was undertaken, making use of CN films prepared in our laboratory.

5.4.2 Experimental

In the fuel pellets under study, the ratio of alpha disintegrations from Uranium to that from Plutonium worked out to 1:2775. This showed that activity-wise the amount of Uranium was so low that it could
be neglected (Bhagwat & co-workers, 1976c). The tracks recorded on GN films during exposure should therefore be considered to be all due to Plutonium. A non-uniform distribution of Plutonium should therefore manifest itself through non-uniform distribution of tracks.

Three films of 1.0" diameter were used for this purpose. These films were individually wrapped in aluminised mylar film of 0.25 mil thickness to avoid their contamination by fuel pellets. Each film was exposed to one fuel pellet in contact with one of the end faces for 5 seconds inside a glove box. After exposure, the films were etched in 6N NaOH at room temperature (24°C) for 4.5 hours, and then examined under the microscope.

Here the recorded track densities of alpha particles were so large that it was not possible to count individual tracks. The data of interest was therefore obtained by looking for clusters or stars of alpha tracks in an otherwise uniform background (of tracks). The autoradiographs reflect only the surface distribution. An important assumption was therefore made that the conditions at the surface are representative of those prevailing in the bulk, and it should be normally so.

All the three films showed the presence of clusters or stars. Some typical microphotographs of the distributions observed are shown in Fig. 5.10 a,b,c. The number of such clusters or stars and their locations on each film were noted. This data was then transferred to graph sheets from which the number of clusters in each unit area of 1 mm² was determined. Fig. 5.11 represents the distribution of these
FIG. 5.10. MICROPHOTOGRAPHS SHOWING (a) A CLUSTER OF 300 μm DIAMETER (b) A GROUP OF 5 CLUSTERS AND (c) AN AREA WHERE THERE ARE NO CLUSTERS.
FIG. 5.11: HISTOGRAMS SHOWING THE NON-UNIFORM DISTRIBUTION OF $PuO_2$ IN $UO_2 - PuO_2$ FUEL PELLETS.
clusters which were found to vary from zero to 6 per mm². The sizes of majority of clusters were found to lie in the range of 30 to 60 μm with a few lying below and above this range.

5.4.3 Conclusion

This investigation not only established the utility of the method but also brought out the fact that complete uniformity in mixing was not attained. The method of mixing of UO₂ and PuO₂ powders was subsequently improved to achieve complete uniformity. This was confirmed by examining additional pellets from subsequent lots whose autoradiographs showed the presence of few or no clusters at all.

5.5 SPONTANEOUS FISSION DECAY CONSTANT (λf) OF 238 URANIUM

5.5.1 Introduction

The experiment was undertaken with a view to build up our confidence in the measurement of low number of events, e.g., fission events, which one may encounter as a part of the routine. One advantage with this measurement was that our value could be easily compared with the large number of values available in literature as the largest number of independent measurements have been made on 238 U (Fleischer and Price, 1964; Roberts & co-workers, 1968; Lom & co-workers, 1973; Khan & co-workers, 1973). Only a few references have been quoted here. Most of the reported values of λf lie in the range 6.8 - 7.5 x 10⁻¹⁷ yr⁻¹ (Fleischer & co-workers, 1975).

5.5.2 Method

The track density Fₚ (no.cm⁻²) in the detector foil, when it is
exposed in contact with a Uranium (natural) sheet/foil, due to spontaneous fission (S.F.) of $^{238}$U atoms is given by (Fleischer and Price, 1964):

$$P_S = (e^{-\lambda_D T} - 1) N c_{238} R \frac{\lambda_F}{\lambda_D} \quad \ldots \ldots (1)$$

where

- $\lambda_D$ = total decay constant for $^{238}$U
- $\lambda_F$ = S.F. decay constant for $^{238}$U
- $T$ = time of exposure
- $N$ = No. of atoms of Uranium.cm$^{-3}$ in Uranium sheet
- $c_{238}$ = fraction of Uranium atoms that are $^{238}$U
- $R$ = effective etchable range of fission fragments in Uranium sheet in cm.

The necessity to determine $N$, $R$ and $c_{238}$ can be avoided by exposing the same sheet of Uranium in contact with another detector foil to a known fluence of thermal neutrons. The new track density $P_T$ due to thermal fission of $^{235}$U is given by:

$$P_T = N c_{235} \sigma \cdot R \quad \ldots \ldots (2)$$

where

- $\sigma$ = fluence of thermal neutrons
- $\sigma$ = thermal fission cross-section of $^{235}$U
- $c_{235}$ = fraction of U atoms that are $^{235}$U.

Taking the ratio of (1) and (2) we get

$$\frac{P_S}{P_T} = \frac{(e^{\lambda_D T} - 1) c_{238} \lambda_F}{c_{235} \sigma \cdot R \lambda_D} \quad \ldots \ldots (3)$$
If, $T < 10^8$ years, $(e^{-123} - 1)$ becomes $\lambda_{D}^{T}$, and denoting $c_{235}/c_{238}$ by $I$, we get,

$$\frac{P_S}{P_I} = \frac{T \cdot \lambda_F}{I \cdot \phi}$$

or

$$T = \frac{P_S}{P_I} \cdot \frac{I \cdot \phi}{\lambda_F} \quad \ldots \quad (4)$$

Eqn. (3) or (4) can be used to determine $\lambda_F$ if $T$ is known.

5.5.3 Experiment and Result

The track density $P_S$ was measured by placing the Makrofol N track detector foil in contact with a polished Uranium natural pellet (1 mm thick) and accumulating the tracks due to spontaneous fission of $^{238}\text{U}$ over a period of several months. Two such pellet-detector combinations were wrapped in 1 mm thick Cadmium foil and stored in an area where there were no known radioactive sources (i.e., neutron sources) of any kind (in the basement of BARC Hospital at Anushaktinagar, Bombay). The irradiation of Makrofol pieces was continued for 2 years and 49 days ($= 2.134$ years = $T$). The detector foils were then separated, etched in 6N $\text{NaOH}$ at $60^\circ\text{C}$ for 1 hour and scanned under the microscope. The value of $P_S$ obtained was $1565 \pm 7 \%$ track/cm$^2$ (203 tracks were counted in 422 views, area of each view being $0.03074 \text{ mm}^2$).

The track density $P_I$ was measured by placing one of the Uranium foils in contact with another Makrofol foil and irradiating the combination with a known fluence of thermal neutrons. The irradiation
was carried out in the standard thermal neutron facility of Radioactive Standardisation Section at Trombay, fluence being $\Phi = 5.99 \times 10^8 \text{n.cm}^2$.

The detector foil after separation was etched and scanned in the fashion mentioned above. The value of $P_I$ obtained was $(28690 \pm 4.8\%) \text{track.cm}^{-2}$ (436 tracks were counted in 40 views, area of each view being $0.03799 \text{ mm}^2$. This was a different microscope).

Taking the value of $\beta$, the fission cross-section of $^{235}\text{U}$ as $5.82 \times 10^{-22} \text{ cm}^2$ and that of I as $\frac{1}{137.8}$ for natural Uranium, the value of $\lambda_f$ from equation (4) was found to be

$$\lambda_f = (6.46 \pm 0.55) \times 10^{-17} \text{ year}^{-1}$$

which is quite close to most of the reported values, which lie in the range $6.8 - 7.5 \times 10^{-17} \text{ year}^{-1}$. Scanning of larger areas may possibly lead to some improvement in the value.