LIST OF CANDIDATE'S PUBLICATIONS


2. Some applications of plastic track detectors and a possible correction for the spark counting of fission tracks. I Asian Regional Congress on Radiation Protection, Bombay, 1974, A.M. Bhagwat, V.R. Deo, Hari Singh and S.D. Soman.


5. Correction for the observed track-hole densities in the spark counting of fission tracks - Nuclear Instruments and Methods, 138, 173 (1976), A.M. Bhagwat, Hari Singh and S.D. Soman.


9. "Applications of SSNTD to radiation dosimetry" - review talk delivered at Seminar-cum-Workshop on SSNTD, Bombay, 1979, A.M. Bhagwat (This talk has been published in IARP Bulletin of Radiation Protection), 2, (no.1), 19, 1979.


* Reprints of these papers are attached. They are arranged in the order in which their contents appear in the thesis.
CORRECTION FOR THE OBSERVED TRACK-HOLE DENSITIES IN THE SPARK COUNTING OF FISSION TRACKS

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The spark counts (hereafter referred to as observed value) for fission-fragment track-holes in thin polycarbonate detector films need a correction to obtain the true value, i.e. microscope counts. A linear relationship has been found to exist between percentage correction (to be added) and the observed track-hole densities up to \( \sim 1000/\text{cm}^2 \) where the magnitude of this correction becomes of the order of 10%. A similar relationship has also been obtained theoretically on the basis of average effective area of a hole on the aluminised mylar film and the probability of loss of counting of subsequent track-holes due to this finite area. The close agreement between these two relationships justifies the method developed in arriving at this correction for the counting losses. The effect of certain factors such as thickness of aluminium layer, capacitance of the circuit and sparking voltage on the size of evaporated aluminium spots has also been studied to establish optimum experimental conditions.

1. Introduction

During the standardisation of a spark counter, based on "Svensson Circuit"\(^1\), for counting fission-fragment track-holes in thin Makrofol (thickness \( = 12 \mu \text{m} \)) it was observed that for any given exposed film the spark count was always smaller than the microscope count, i.e. losses occur during spark counting. Although such losses, which are significant in regions of high track densities have been reported\(^2,3\) to be due to overlapping of the evaporated aluminium spots, correction for obtaining the true count, i.e. microscope count, has not been suggested. We have made an effort in this direction and suggest corrections due to overlapping of evaporated aluminium spots. Factors affecting this spot size of evaporated aluminium are also investigated and discussed.

2. Experimental

In these experiments, we have used 12 \( \mu \text{m} \) thick polycarbonate film (Makrofol) and the radiator consisted of a thick pellet of natural uranium of area 2.453 cm\(^2\). To make the detector films suitable for

![Fig. 1. Holes in the aluminium replica covering (a) one (normal) hole; (b) two or more holes in the detector film.](image-url)
These are shown as continuous lines in the figure. The percent coefficient of variation of spark counts is approximately 0.00314 \(X\) (for \(X = 200\), it amounts to 0.63\%).

A similar relationship has also been obtained on the basis of the average effective area of a hole on the aluminised mylar film and the probability of loss of counting of subsequent track-holes due to this finite area. This is outlined below:

The average effective area of a hole in the aluminised mylar film has been determined by:

1) Using the formula \(\frac{1}{2} \pi d_1 d_2\) for the area of the hole, because the holes are not exactly circular (\(d_1\) represents the largest hole diameter and \(d_2\) the diameter perpendicular to it).

2) Taking five representative holes from aluminium replica of each detector film to make the area value as representative as possible.

3) Using the formula \(\pi (r_1 - r_2)^2\) to calculate the effective area of the hole. This effective area is shown in fig. 3 and can be explained as follows: \(r_1\) represents the average radius of a hole 'A' in the aluminised mylar film; \(r_2\) represents the average radius of a hole 'B' in the detector film which caused the hole 'A'. 'C' is any other hole in the detector film. The probability of loss of counting hole 'C' shall be unity as long as it lies completely within the hole 'A'. Thus the effective average area of hole 'A' which would be responsible for the loss of counting of hole 'C' is the area with radius \((r_1 - r_2)\). Let \(\pi (r_1 - r_2)^2 = a\). Now let us assume that,

\[N_k = \text{number of events on the detector film per unit area,}\]
\[N_s = \text{number of events detected by the spark counter per unit area,}\]

and

let \(f(n, a)\) be the total effective area occupied when \(n\) events have occurred per unit area.

Then, because the function \(f\) also gives the probability of loss of a forthcoming event, it must satisfy the recurrence relation.

\[f(n + 1, a) = [1 - f(n, a)] a + f(n, a).\]  \(\tag{4}\)

The form of \(f\) can be easily obtained by following the events one by one and taking \(f(1, a) = a\). The form of \(f\) thus found is

\[f(n, a) = \sum_{r=1}^{n} (-1)^{r+1} nC_r a^r.\]  \(\tag{5}\)

Relation (5) can be proved by the method of induction and it satisfies relation (4).

Now, before we can equate \(N_k\) with \(f(N_k, a)\), a word of caution is necessary. According to this if \(n\) events have occurred, then \(f(n, a)\) is the expectation value of numbers to be seen but the converse is not strictly true. However we assume that the discrepancies are negligible particularly when \(n\) is not too small. With this comment we write

\[N_k a = f(N_k, a) = 1 - (1 - a)^{N_k}.\]

\[N_k = \frac{\log(1 - N_k a)}{\log(1 - a)}.\]  \(\tag{6}\)

Expanding the log terms and neglecting terms containing \(a^2\) and higher order of \(a\), relation (6) can be approximated to

\[N_k = N_k (1 + \frac{1}{2} N_k a).\]  \(\tag{7}\)

Now since \((N_k - N_k)/N_k X 100\) is represented by \(Y\) and \(N_k\) by \(X\), we get

\[Y = 50 a X.\]  \(\tag{8}\)

With \(a = 1.8613 \times 10^{-4}\ \text{cm}^2\), the theoretical line of best fit is

\[Y = 0.009307 X,\]  \(\tag{9}\)

which is in close agreement with relation (1) based on experimental data.

Our discussion thus suggests that the true track-hole density can be obtained from the observed density \((X)\) (given by the spark counter) and is equal to \(X + \frac{1}{2} a X^2\), where \(a\) is the average effective area of a hole in the aluminised mylar film. The value of \(a\) can be determined for the system once for all (unless the system is modified) and the correction applied accordingly.
the counting error. But it was further observed that below \( \sim 100 \text{ pF} \), although the size of holes formed continues to decrease, the spark counts obtained show a dependence on the type of scaler used. A valve type utility scaler (dead time \( \sim 250 \mu s \)) gives a lower value of spark counts than an I.C. scaler (dead time \( \sim 1 \mu s \)). Since the I.C. scaler is not easily accessible at present, we have decided to use a value of 100 pF for the time being. Studies on the effect of these and other parameters have also been reported by Nishiwaki et al.\(^1\).

5. Conclusion

The close agreement between the experimental and the theoretically developed relationships justifies the method of correction for counting losses. But the actual relationship will have to be developed for each type of spark counter because the size of aluminium spots formed depends on various parameters which must be optimised as discussed above.

References

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A SIMPLE SET-UP FOR SINGLE SURFACE ELECTROCHEMICAL ETCHING (ECE)
OF CHARGED PARTICLE TRACKS

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A large scale amplification of charged particle tracks is achieved by electrochemical etching of SSNTD films. The use of a simple electrodeposition cell for this purpose is described. The advantages which it offers over the setups in use in other laboratories, are also described.

The recent work\(^{(1-3)}\) on electrochemical etching for the amplification of charged particle tracks has demonstrated that the technique has a promising future, especially in the field of radiation dosimetry. The technique is superior to conventional chemical etching because it permits such a large scale amplification \((2 \times 10^4\) for tracks of \(200 \mu\)m diameter, assuming an original latent track diameter of \(100 \AA\)) of the tracks that they can be either seen by naked eye or can be projected on a screen with a slide projector. To take advantage of this fact, a simple ECE setup, described below, has been used in our laboratory for routine use.

An exploded view of the ECE cell, which is normally used in electrodeposition experiments, is shown diagramatically in fig. 1. The exposed side of the film faces the high voltage electrode. If the tracks can appear on both sides, as is the case with recoil atom tracks from neutron irradiation, that side will face the high voltage electrode on which the track development is desired. The contact between the film and the ground electrode is improved by introducing a drop of water in between. This improves the quality of tracks.

The high voltage function generator used in our experiments is a combination of a function generator and a transformer with a step-up ratio of 30. It is capable of giving a sinusoidal output from a few volts to 1800 V (peak-to-peak) in the frequency range 1-10 kHz. Its current capacity is about 3 mA.

Lexan\(^*\) polycarbonate film of \(280 \mu\)m thickness was employed in the experiments with this setup. This film is protected on one side with a thin plastic sheet. This protective sheet is removed only at the time of exposure/ECE as the case may be. This surface is therefore practically free from scratches. The background track density in most of these films is around 2-3/cm\(^2\). This background may be due to the presence of foreign matter or faults (microvoids) on the surface. Such flaws disturb the voltage distribution and assist in initiating the treeing process.

Some of the results obtained with the above setup for Lexan \((280 \mu\)m thick) using KOH\((400 \text{ g/l})\) at room temperature \((24-25 ^\circ\text{C})\) are given in table 1.

\* Trade mark of General Electric Co, U.S.A.
over a period of 6 h or more when ECE is carried out at room temperature. This rise in temperature further facilitates track development.

2) The cell permits ECE at higher temperatures also, i.e. up to about 75°C as it can be surrounded conveniently by a small heating jacket for this purpose.

3) Two or more cells can be connected simultaneously, in parallel, to the same function generator.

4) Since only one side of the film is subjected to ECE, the question of background contribution from the other side does not arise. This is a serious drawback in double surface ECE especially when film thicknesses are small.

5) Polycarbonate films with mat finish on one side, i.e. all grades of Makrofol*, can be conveniently used with this setup as it is possible to avoid ECE on the side with mat finish. In double surface ECE, the side with mat finish develops a large number of background tracks.

The authors wish to acknowledge the help rendered by Ss. J. K. Samuel, M. R. Ponkshe and K. S. Kamraj of the Radiochemistry Division during the course of this work.

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* Trade mark of Bayer A.G., West Germany.
ON THE INVESTIGATION OF SOLUTION (ETCHANT) PARAMETERS INFLUENCING THE DEVELOPMENT OF CHARGED PARTICLE TRACKS BY ELECTROCHEMICAL ETCHING (ECE)

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ABSTRACT

Conductivity, viscosity and track-etch rate measurements were carried out for KOH solutions of various normalities and their mixtures (50% v/v) with ethyl alcohol (hereafter referred as alcohol). The type of tracks studied were fast neutron induced recoil atom tracks and fission tracks in Lexan. It was observed that the presence of alcohol does not improve the EC track etch rates as significantly as it does for chemical track etch rates. This has been explained as follows: Alcoholic solutions have lower conductivity (in addition to higher viscosity). This suppresses to some extent the favourable effect of alcohol.

EC track etch rates (i.e. diameter measurements) show a saturation at KOH concentrations of about 7 N, similar to chemical track etch rates (length measurements) which have a peak around 12 N. While the peak in chemical etching (CE) is due to viscosity alone, the same in ECE is due to both viscosity and conductivity. This shift of peak position in ECE towards lower concentrations is a result of lower conductivities at higher concentrations. It was also observed that very small volume (less than 0.5 ml) of etchant is adequate for ECE of areas about 3 cm² large. Because of this smaller requirement of etchant volume, it was possible to modify the ECE cell further to make it simpler.

KEYWORDS

Electrochemical etching; fission tracks; recoil tracks; viscosity; conductivity;

INTRODUCTION

The phenomenon of ECE of charged particle tracks has recently been a subject of intense study in a number of laboratories (Al-Najjar, Bull and Durrani, 1979; Hassib and Fleisch, 1978; Schrabi and Morgan, 1978; Somogyi and others, 1979; Tommasino and others, 1979a, 1979b) around the world. This is primarily because of large scale amplification of latent tracks which the technique affords. Studies on this technique were initiated in our laboratory some time ago mainly to explore its utility for personnel fast neutron dosimetry. This led us to successful development of single surface ECE technique using an Electrodeposition cell. Our initial results
on this technique have already been reported (Bhagwat and Soman, 1979). An effort was since then made to study systematically the influence of various experimental parameters relevant in ECE. Of these, the studies on solution parameter are reported here.

CONDUCTIVITY AND VISCOSITY MEASUREMENTS

Since alcoholic solutions of KOH lead to faster development of tracks in both CE and ECE, effect of alcohol was first studied on the conductivity of KOH solutions. It was observed that, with the addition of increasing amounts of alcohol, the conductivity first dropped rapidly and then slowly, the drop being very gradual when the volume of alcohol added approached the volume of KOH solution (it was therefore decided to take alcohol and KOH solutions in equal volume proportion while using mixtures). Since, alcoholic solutions give higher EC etch rates, it indicates that conductivity is not the sole deciding factor.

Viscosity measurements showed that viscosity of alcoholic mixtures is higher than that of pure KOH solutions. The increase in viscosity is due to the fact that alcohol water mixtures in any proportion have higher viscosity than either pure water or pure alcohol. The mixing is also associated with slight reduction in volume. We also know that higher viscosity is associated with lower etching rates (compare KOH and NaOH solutions of equal normality). Thus to explain the higher etching rates of alcoholic solutions, additional investigations must be made.

ROLE OF ALCOHOL

As conductivity and viscosity measurements did not explain adequately why the presence of alcohol enhances EC etch rates, a few etch rate measurements, given below, were made to understand the role of alcohol.

Bulk chemical etch rate measurements carried out earlier for Lexan film (Bhagwat and Soman, 1981), showed that pure alcohol produces no etching at all. On the contrary, Lexan gains weight in alcohol as a result of swelling. This swelling causes relaxation of secondary valance forces between long linear polymer chains and enhances their susceptibility to chemical attack.

A few diameter measurements, given below, were also made to understand how the presence of alcohol influences them (values are for normally incident fission fragments in Lexan, etching temperature being 24°C). This data is also represented in

\[
\begin{align*}
&i) \quad \text{with } 6 \text{ N KOH} \quad \text{CE} \quad 0.162 \text{ µm/HR} \\
&ii) \quad 6 \text{ N KOH + alcohol} \quad \text{CE} \quad 2.74 \text{ µm/HR} \\
&iii) \quad 6 \text{ N KOH} \quad \text{ECE} \quad 22.8 \text{ µm/HR} \\
&iv) \quad 6 \text{ N KOH + alcohol} \quad \text{ECE} \quad 32.5 \text{ µm/HR}
\end{align*}
\]

Fig. 1. It helps us to clearly identify the role of alcohol in the two processes. Thus in ECE, the track etch rate enhancement achieved due to presence of alcohol i.e. about 14 is very much lower when compared with an enhancement factor of about 16.9 obtained in case of chemical etching. The lower enhancement in ECE is due to lower conductivity and higher viscosity of alcoholic solution, coupled with track shape as described below.

SATURATION EFFECT IN EC TRACK ETCH RATES

Saturation effect has been reported by Sohrabi (1975) and observed by us also in
EC track etch rates (with varying concentration of KOH solution). As no explanation was available for the same, this was investigated in somewhat greater detail. Bulk etch rates and track etch rates (based on diameter measurements) in CE show no saturation for Lexan for both pure and alcoholic KOH solutions. This is because both these measurements represent a surface phenomenon. Chemical etch rates were therefore measured based on observed track lengths. This data represented in Fig. 2, shows existence of a peak around 12N. We explain this peak as follows:

In CE, the rate at which the etchant penetrates the track region decreases as its viscosity (i.e. concentration) increases. However, the etching power of the solution increases with concentration. The situation at the solid-liquid interface "inside the track region" is therefore very typical. At this interface the etchant concentration drops due to attack on the polymer and the high viscosity of the etchant does not permit its quick replacement by fresh solution from the bulk. As a result there is a drop in track etch rate. This situation, which can be considered as a "shape or Geometry factor" gets further magnified in ECE due to voluminous nature of the track. Here a large track is connected to the surface (Griffith and others, 1977) through a small channel or orifice. During ECE therefore, it can be visualised as though a small reservoir (ECE track) is connected to a big reservoir (bulk of etchant) through the small opening or the orifice. At high viscosities, this small orifice does not permit rapid mixing of solutions in the two compartments and leads to the observed effects.

The peak etch rate in ECE corresponds to a concentration of about 7N. This shift towards lower concentration (as compared to peak position in CE) is due to 1) conductivity saturation for etchant occurring at lower concentration and 2) the small opening of ECE track which enhances the effect of viscosity.
Fig. 3. Simplified ECE setup.

VOLUME OF ETC HAFT REQUIRED

ECE experiments were conducted using 8.0, 4.0, 2.0, 1.0 and 0.5 ml of etchant (10N KOH + alcohol). In all these cases the track sizes were essentially the same. In the light of this experience, a new cell arrangement, shown in Fig. 3, was tried and found useful. Here the high voltage electrode also acts as a container/holder of the etchant. The etchant volume held depends upon the depth of the electrode cup. Its biggest advantage is that the film mounting procedure is very simple. One need not worry about the problem of leak tightness, as long as the film is slightly bigger than the electrode faces.

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ON THE ROLE OF ALCOHOL AND ELECTRIC FIELD IN THE
ETCHING PROCESSES - CHEMICAL AND ELECTROCHEMICAL

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I. Chemical etching

Polycarbonate film which is being commonly used in both chemical and Electrochemical etching (ECE) studies was selected for this investigation. The etchants consisted of (i) 6N KOH soln. (ii) an equal volume mixture of 6N KOH and ethyl alcohol and (iii) ethyl alcohol alone, etching temperature being 60°C.

For etchants (i) and (ii) the etching rates observed were 146 and 1566 µg/h/cm² respectively. But for the third etchant, i.e., pure ethyl alcohol, no etching effect was observed. On the contrary there was weight gain to the extent of 42 µg/h/cm².

In the light of these results, the role of ethyl alcohol can be interpreted as follows - The presence of ethyl alcohol produces swelling of the Polycarbonate foil by penetration which in turn causes relaxation of secondary valance forces between long linear polymer chains and enhances interchain separation. This not only increases the susceptibility of separated polymer chains to chemical attack but also enhances penetration.
of the etchant as a whole.

In case of etching with pure alcohol, the weight gain disappeared after some time indicating thereby that retention of alcohol is not permanent. In case of etching with alcoholic mixture, there was no further loss of weight on retention indicating that alkali action on separated (loosened) chains is quick which does not leave any scope for alcohol remaining trapped after etching has ceased.

II. Electrochemical etching

From the data (see Fig. 1) on track diameter growth versus time of ECE for fast neutron induced recoil atom tracks (conditions of ECE : 2 kVp, 2 kHz, etchant = equal volume mixture of 10N KOH and C₂H₅OH) it is clear that etching rate initially increases with time and then becomes constant. For the recoil tracks as well as fission fragment tracks encountered in routine dosimetry work we are dealing with damage density regions at low energy end of the curves for damage density versus energy/amu of the charged particles (Fig. 1-8, Page 18, ref. 1). This is magnified and shown in Fig. 2. In this region damage density falls off as the particle, producing track, slows down. The shapes of chemically etched tracks corresponding to this region are similar to that shown in fig. 2-10, p.63, No.1, ref. 1, and reproduced in fig. 3. This figure clearly establishes the point that as the chemical etching proceeds the ratio of major axis to minor axis decreases or the tip radius 'r' increases or the distance 'd' of the tip from the other surface decreases.
Now let us look at the following equations (1) and (2):

\[
\frac{E_{\text{max}}}{E_{\text{av}}} = 2n^{\frac{3}{2}} m \ln \left( \frac{m-n}{m-n} \right) - 2n \quad \text{.... (1) (By Bateman) (ref. 2)}
\]

where \( E_{\text{max}} \) = electrical field at the tip of the track

\( E_{\text{av}} \) = average electrical field applied to insulator

\( m \) = ratio of major axis to minor axis = \( c/a \)

\( n = \sqrt{m^2 - 1}, \quad \text{AND} \)

\[
\frac{E_{\text{max}}}{E_{\text{av}}} = \frac{2d}{r} \ln \left( 1 + \frac{4d}{r} \right) \quad \text{.... (2) (By Mason) (ref. 2)}
\]

where \( E_{\text{max}} \) & \( E_{\text{av}} \) are the same as in eqn. (1)

\( r \) = radius of the tip

\( d \) = distance of the tip from opposite surface of film.

These equations give electrical stress at the tip of the track. During ECE, chemical etching changes 'c/a' or 'd' or 'r' in a way as to reduce the electrical stress at the tip of the track. Hence EC etch rate should drop as ECE proceeds. Since this will be contrary to observations, it should be possible to explain the experimental results as follows: -

During ECE, changes in "c/a" or "d" or "r" do not keep pace with the advancing track tip with the result that at the tip the conditions are always conducive to further growth. Thus in ECE with alcoholic KOH solutions -
Electric Stress produces and propagates cracks with sharp tips; alcohol produces swelling action and both of these in turn facilitate chemical action. The importance of sharp tips becomes evident when we consider that no ECE is possible for chemically etched tracks where tips have become rounded.

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FIG. 1. TRACK DIAMETER VS EC ETCHING TIME FOR A RECOIL ATOM TRACK IN LEXAN (130 μm THICKNESS; 2 kVp; 2 kHz; 10 N KOH + C₂H₅OH)
FIG. 2. VARIATION OF ELECTRONIC STOPPING POWER (OR DAMAGE DENSITY) VERSUS ENERGY/α µm FOR VARIOUS CHARGED PARTICLES IN MYLAR (POLYETHYLENE TERPHTHALATE, C₁₀H₈O₄).
FIG. 3 PROFILE OF A CHEMICALLY ETCHED TRACK WITH ETCHING TIME, WHERE DAMAGE DENSITY DECREASES WITH DEPTH.
A STUDY OF THE ROLE OF ELECTRICAL PARAMETERS IN THE ELECTROCHEMICAL ETCHING (ECE) PROCESS

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ABSTRACT

The effect of frequency on the density and size of fission tracks and fast neutron induced recoil atom tracks was studied. It was observed that while the choice of frequency is wide (from less than 1 kHz to 4 kHz) for highly ionizing particles like fission fragments, low frequencies (0.25 - 0.5 kHz) are better suited for recoil atom tracks. The differences observed between normally incident fission tracks and recoil atom tracks are probably due to the fact that the latter tracks have a spectrum of energies (extending to almost zero energy at the lower end) and secondly that they lie at all angles in 2π geometry. Alcoholic solutions (50% v/v) produce higher densities of recoil atom tracks and give rise to higher background too. Higher voltages at any given frequency give improved results and hence are preferred.

When the above data on the effect of frequency was compared with data on Leakage Current (IC) versus frequency, no apparent relationship appeared to exist. However, when two films of different thicknesses were studied under identical conditions of ECE (for normally incident fission fragments) i.e. at constant frequency of 1 kHz and one of the following conditions: same total voltage or IC or Vp/μm or IC/μm, it was observed that for the same total IC, tracks of nearly the same sizes were obtained. This suggests that the total IC should be an important parameter in any further investigations.

KEYWORDS

Electrochemical etching; fission tracks; recoil tracks; leakage current; frequency.

INTRODUCTION

The acceptability (and usability) of Solid State Nuclear Track Detection (SSNTD) technique has been greatly enhanced by various methods of track visualisation and evaluation introduced over the past one decade or more. ECE technique which provides large scale amplification of charged particle tracks, is one such method of easy track visualisation and their consequent evaluation. Having introduced the ECE technique in our laboratory (Bhagwat and Soman, 1979) for fast neutron
dosimetry, a systematic study of various electrical and solution parameters became essential for a better understanding of this process. Investigations of electrical parameters are described here. Studies on solution parameters by the same authors (Bhagwat and Soman, 1981) are reported elsewhere in this conference.

EFFECT OF FREQUENCY AND VOLTAGE

This was studied for both track density and track sizes, for recoil tracks in Lexan, using pure KOH solution and alcoholic KOH solution (50% v/v). It was observed that the track density increased as the frequency of applied voltage decreased. A typical result is shown in Fig. 1, for pure KOH solution. Control films also showed a similar trend as can be seen from this Fig. The track densities, for both irradiated and control film were larger when alcoholic solution was used. It is thus clear that the net track density increases as the frequency decreases. The sensitivity i.e. tracks/neutron, also increases in a parallel fashion.

The above curves could not be studied further towards the lower frequencies because of limitations on equipment output. However, these curves cannot be expected to rise indefinitely as they must drop at some stage to permit them to pass through origin. Thus it appears that the peak lies somewhere between zero and 0.25 kHz. This position is different from that reported by Sohrabi (1975) which is in the region of 1.5 to 2.0 kHz. We have no immediate explanation for this observed difference except that it may be a result of different circuit parameters in the two cases.

For alcoholic solutions, the size (maximum average diameter) versus frequency curve showed a broad peak at 1.5 kHz. Similar peak was not observed for pure KOH solution. This was partly due to the fact that very few tracks were obtainable above
1.0 kHz for the irradiations carried out. This indicates that the effect of frequency on track sizes is not as significant as on track density. Taking the overall view, the frequency range of 0.25 to 0.5 kHz appears to be ideal for pure KOH solutions. For alcoholic solutions, the frequency will also depend on the proportion of background tracks acceptable. In the light of this limitation the range of 0.5 to 1.5 kHz may be considered suitable for the latter case.

For the effect of voltage on track density, at a given frequency, it was observed that up to a voltage of 2000 Vp (the maximum obtainable with our equipment), there is substantial improvement in net track density with increasing voltage. The effect of time of ECE was also considered. When we compare the number of tracks obtained at 90 and 160 minutes of ECE (for recoil atom tracks, using alcoholic KOH solution at 1.0 kHz and 2kVp), there was no significant gain in net number of tracks, although background increased tremendously. Thus the time of ECE should be kept minimum consistent with track size requirement, though longer etching times produce larger tracks which are easy to count.

COMPARISON WITH EC ETCHED FISSION TRACKS

When the effect of frequency on the ECE of normally incident fission fragment tracks was considered, it was observed that frequency had no effect on track density in the range from less than 1.0 kHz to 4.5 kHz. This difference in the effect of frequency on recoil atom tracks and fission tracks may be due to following reasons: i) recoil tracks have a spectrum of energies (extending to almost zero on the lower energy side) and ii) recoil tracks lie in Z geometry. It thus appears that lower frequencies are better suited for tracks having angular and energy distribution similar to that of recoil tracks.

LEAKAGE CURRENT MEASUREMENTS

A possible role for IC, in the development of ECE tracks has been mentioned in literature (Schrab, 1975; Johnson and others, 1978). This parameter was also therefore investigated. It was observed that for various films such as Lexan, Makrofol, CTA, CN and local polyester films, the pattern of IC versus frequency curves was similar for both unirradiated and irradiated (with gamma dose of 50 to 100 Mrad) films. A typical set of curves is shown in Fig. 2. These curves show a very broad maximum (peak) in the region of 3 to 4 kHz and have no apparent resemblance with frequency versus track density curves for recoil tracks and fission tracks. However, the differential IC (difference in values for the irradiated and unirradiated films) has a maximum, though not sharp around 0.5 to 0.7 kHz for all the films. It was therefore expected that this maximum should manifest itself in some way during the ECE process.

A comparison was therefore made between Lexan - a film which responds nicely to ECE and CTA - a film which responds poorly to ECE. It was observed that in Lexan no effect was noticed which could be assigned to the observed maximum in the differential IC. However, for CTA the best results that could be obtained corresponded to the region of this maximum. It may thus be concluded that the maximum in differential IC has only a minor role to play which may manifest itself when other factors for track growth are not very favourable. It was further observed that at any given frequency, the IC increases linearly with voltage (up to 2000 Vp). Thus once a frequency is chosen, higher voltages will give faster track development as stated earlier.
EFFECT OF FILM THICKNESS

The purpose of these experiments was to look for a parameter which controls ECE best irrespective of thickness variations. Accordingly, Lexan films of two different thicknesses were chosen, i.e., 400 µm and 240 µm. They were exposed to normally incident fission fragments and subjected to ECE at constant frequency (1.0 kHz) and one of the following conditions:

i) same total voltage across the film
ii) same total LC through the film
iii) same Vp/µm of film thickness across the film
iv) same LC/µm of film thickness through the film

The basis for comparison of results was track diameters (as all the normally incident fission fragments grow into ECE etched tracks under the specified experimental conditions). The results obtained are shown in Table 1. From these experiments, it is seen that when total LC is kept constant, tracks of very nearly the same size are obtained. The parameter that can be considered next best is constant Vp/µm. This suggests that the total LC is quite an important parameter (in addition to frequency) and should form an integral part of any further investigations.

**TABLE 1  Effect of Thickness on Diameter Growth**

<table>
<thead>
<tr>
<th>S.no. of film</th>
<th>Film Thickness</th>
<th>Parameter kept constant</th>
<th>Track diameter</th>
</tr>
</thead>
<tbody>
<tr>
<td>(1) and (2)</td>
<td>403 µm</td>
<td>LC/µm</td>
<td>90.04 µm</td>
</tr>
<tr>
<td>(3) and (4)</td>
<td>247 µm</td>
<td>0.446 Vamp/µm</td>
<td>71.7 µm</td>
</tr>
<tr>
<td>(1) and (2)</td>
<td>403 µm</td>
<td>total LC</td>
<td>90.04 µm</td>
</tr>
<tr>
<td>(5) and (6)</td>
<td>239 µm</td>
<td>180 Vamp</td>
<td>95.27 µm</td>
</tr>
<tr>
<td>(7) and (8)</td>
<td>405 µm</td>
<td>Vp/µm</td>
<td>98.4 µm</td>
</tr>
<tr>
<td>(9) and (10)</td>
<td>248 µm</td>
<td>5 Vp/µm</td>
<td>89.32 µm</td>
</tr>
<tr>
<td>(7) and (8)</td>
<td>405 µm</td>
<td>total Vp</td>
<td>98.4 µm</td>
</tr>
<tr>
<td>(11) and (12)</td>
<td>236 µm</td>
<td>2025 µm</td>
<td>118.35 µm</td>
</tr>
</tbody>
</table>

*For fission tracks at 1.0 kHz using 10N KOH + C,H, OH for 90 min in Lexan.

CONCLUSION

From the above studies, it is abundantly clear that the right choice of electrical parameters is very important to obtain optimum results. Each type of film must therefore be investigated in the above fashion before use. The role of solution parameters is also equally important and they must be considered together with electrical parameters in any optimization process.

REFERENCES

ON THE ROLE OF POLARISATION AND POLYMER TYPE IN THE PROCESS OF ELECTROCHEMICAL ETCHING (ECE)

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Health Physics Division
Bhabha Atomic Research Centre
Trombay, Bombay 400 085

ABSTRACT

The effect of polarisation and the polymer type on the process of ECE is examined. It is suggested that dipolar polarisation alone plays an important part as far as the film material is concerned. As regards the polymer type, it is concluded that the polymers should also have i) a low value of dissipation factor or dielectric loss factor and ii) polar nature.

INTRODUCTION

Electrochemical Etching (ECE) method of visualising charged particle tracks in polymers has gained wide acceptance in recent years primarily because of the fact that it simplifies the subsequent job of track counting to a great extent. It is achieved through large scale amplification of the original latent tracks. Various parameters which influence the process of ECE have been investigated by us and many others (Bhagwat and Soman, 1981a, 1981b; Reali and Burrani, 1981; Al-Hajjar and Co-workers, 1979; Johnson and Co-workers, 1978; Soregyi, G., 1977; Sohrabi, M. 1975) and reported in literature. Their importance in establishing the optimum conditions of ECE cannot be underestimated.

In this paper an effort is made to explain the role played by two parameters, i.e. Polarisation and Polymer Type, in more detail.

EFFECT OF POLARISATION

It is known that the power (energy) transferred to an insulating
material, when subjected to an alternating field is a function, not only of the potential across the material and its frequency but also of the physical characteristics of the material. It is therefore understood that the properties of the material also influence the energy transferred.

One of the important effects of the electric field on the insulating material is to displace the bound charges in it slightly relative to one another. Each molecule then acquires an electric dipole moment parallel to and in the same direction as the electric field. This effect is known as dielectric polarization.

There are four distinct mechanisms which contribute to the process of polarization: (i) electronic, (ii) atomic (ionic), (iii) dipolar and (iv) interfacial. Each of these processes is characterized by a relaxation time $T$ which is the time taken by the particular polarization to respond to the applied electric field. The polarization times for the above four processes are: (i) $10^{-16}$ sec, (ii) $10^{-12} - 10^{-13}$ sec, (iii) $10^{-3} - 10^{-10}$ sec and (iv) $1 - 10^{-3}$ sec respectively. For any polarization to make its full contribution to total polarization, the period of oscillation $T = \frac{1}{f}$ of the field (where $f$ is the frequency) should be much greater than the corresponding relaxation time $T$, so that the polarization is able to establish itself completely before the field is reversed (Birks, 1960).

In most of the studies on EOE, one is dealing with dielectric materials which are homogeneous in nature. The interfacial polarization should then be of no consequence as it applies only to inhomogeneous dielectrics. Even if dielectrics under investigation have inhomogeneities of any kind, it may be remembered that interfacial polarization will be extremely slow in establishing itself ($1 - 10^{-5}$ sec) as mentioned above. Very often it is so
slow that it takes place at frequencies which are very much lower than 1 Hz (Barrop, 1972). We also know from the experimental results that the frequencies in the range of 0.5 to 50 kHz make a tremendous difference in the results of BEB. Since this frequency range will affect only the dipolar polarisation it may be considered that this type of polarisation alone plays an important part in the process of BEB as far as the track recording material is concerned. By implication it means that the role of electronic and atomic (ionic) polarisations is small.

EFFECT OF POLYMER TYPE

The table (1) lists a number of properties of seven different film materials, e.g. Cellulose Acetate (CA), Cellulose Tri-Acetate (CTA), Cellulose Acetate-Butyrate (CAB), Polycarbonate (PC), Polyethylene Terphthalate (PET), Polyethylene (PE) and Poly Tetra Fluoro Ethylene (PTFE), for which data was available in literature (British Plastics Year Book, 1972, Modern Plastics Encyclopedia, 1979-80). Of these, the effects of first four properties i.e. tensile strength, dielectric strength, volume resistivity and water absorptivity were discussed by Johnson and Co-workers (1977) for the first four materials, i.e. CA, CTA, CAB and PC. Some of the values given in our table are different from those given by Johnson. As stated above, our values refer to films since we are dealing with films only. (The form of material has not been specified by Johnson in her table). We have added three more materials to the list, i.e. PET, PE and PTFE. Further, a few more properties like dielectric constant, dissipation factor and the dielectric loss factor were considered for all the materials.

The picture now appears to have changed a little. Based on tensile strength, dielectric constant and volume resistivity, PET appears to be superior
### Table 1

**Properties of plastic films**

<table>
<thead>
<tr>
<th>Material</th>
<th>Tensile strength p.s.i.</th>
<th>Dielectric strength volts/mil</th>
<th>Volume resistivity ohm.cm</th>
<th>Water absorption % for 24 h</th>
<th>Dielectric constant for 1 mil thickness at 1.0 kHz</th>
<th>Dissipation factor for 1 mil thickness at 1.0 kHz</th>
<th>Dielectric loss factor at 1 kHz</th>
</tr>
</thead>
<tbody>
<tr>
<td>CA</td>
<td>7000-16,400</td>
<td>1700-3200</td>
<td>$10^{10}$ - $10^{15}$</td>
<td>3.6</td>
<td>0.013</td>
<td>0.0468</td>
<td></td>
</tr>
<tr>
<td>CTA</td>
<td>9000-16,000</td>
<td>2250-3100</td>
<td>$10^{15}$ - $10^{15}$</td>
<td>3.2-4.5</td>
<td>0.016</td>
<td>0.0512</td>
<td></td>
</tr>
<tr>
<td>CAB</td>
<td>5000-9000</td>
<td>2250-3700</td>
<td>$10^{14}$ - $10^{15}$</td>
<td>2.9</td>
<td>0.013</td>
<td>0.0377</td>
<td></td>
</tr>
<tr>
<td>PC</td>
<td>5400-6800</td>
<td>6300(1 mil)</td>
<td>$10^{16}$</td>
<td>0.35</td>
<td>2.99</td>
<td>0.0015</td>
<td>0.004485</td>
</tr>
<tr>
<td>PET</td>
<td>20,000-35,000</td>
<td>7500(1 mil) 2000(5 mil)</td>
<td>$10^{18}$</td>
<td>$&lt;0.8$</td>
<td>3.2</td>
<td>0.005</td>
<td>0.0160</td>
</tr>
<tr>
<td>PE (low density) 4000</td>
<td>1500-5000</td>
<td>5000(1 mil) 3000(5 mil)</td>
<td>$10^{16}$</td>
<td>$&lt;0.01$</td>
<td>2.2</td>
<td>0.0003</td>
<td>0.00066</td>
</tr>
<tr>
<td>PTFE</td>
<td>1500-4500</td>
<td>1200(1 mil) 1000(5 mil)</td>
<td>$10^{18}$</td>
<td>Nil</td>
<td>2.0-2.1</td>
<td>0.0002</td>
<td>0.0004</td>
</tr>
</tbody>
</table>

References: Modern Plastics Encyclopedia (1979-80); British Plastics Year Book (1972).
to all other materials (only the water absorption is somewhat higher than that for PC) but the track formation in it is not as good as in PC. Thus, while the importance of these properties cannot be underestimated, they do not tell the full story.

When we consider the additional three properties mentioned above, for all the materials, a little more light is shed on the topic (here we confine our discussion to the values of these properties measured at 1 kHz only because that is the point about which most of the experimental values have been determined). At 1 kHz, difference between the highest and the lowest values of dielectric constants for these materials is less than a factor of two, i.e. they are about the same. This property therefore does not bring out the differences significantly. But when we consider the values of dissipation factor or dielectric loss factor, tremendous differences are observed.

Thus, considering the first five materials it is observed that PC, which gives best results for BSE, has a low value of both dissipation factor and the dielectric loss factor. PBT, which shows somewhat higher values of these quantities, gives results that are next best, while CA, CTA and CAB which show quite high values of these quantities, give very poor results in BSE. It may therefore be inferred that a low value of dissipation factor of dielectric loss factor is very much essential for obtaining good results in BSE.

On this basis, PE and PTFE, which show still lower values of dissipation factor or dielectric loss factor, should give better results than those for PC. But this is not the case (In PE it can take several days to produce BSE tracks while no results have been reported for PTFE so far). This anomaly will be understandable when we remember that PE and PTFE are both non-polar materials.
Thus, it should be possible to conclude that good ECE tracks should result in materials with the following properties -

i) good electrical properties, i.e. high values of tensile strength, dielectric strength and volume resistivity.

ii) low water absorptivity.

iii) low value of dissipation factor or dielectric loss factor.

and iv) polar nature.

The last criterion, i.e. polar nature of the material is especially important because it has a major role to play in dipolar polarization. The importance of polarization and hence the polymer type is thus obvious.

CONCLUSION

It is thus seen that both polarization and polymer type have an important role to play in the process of ECE. It may however be remembered that in spite of the above information, we are still far from being able to suggest quantitative criteria for track registration by ECE.

REFERENCES

PASSIVE MEASUREMENT OF RADON AND THORON USING TLD OR SSNTD ON ELECTRETS*

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(Received 23 July 1981; accepted 18 December 1981)

Abstract—An electret is an electrical analogue of a permanent magnet and it carries a permanent electric charge. Our previous work has shown that such electrets are suitable for collecting decay products of radon and thoron in passive chambers. In the present work, the decay products were directly collected on the surface of a TLD or SSNTD providing in situ registration of the radiation from the decay products of radon and thoron. A 101. chamber, the sides of which were covered with a layer of Whatman No. 1 (W-I) filter paper, showed the following responses: (i) SSNTD (CR-39) recorded 92 ± 13 tracks per cm$^2$ per pCi/l. hr for radon and 9 ± 1.5 tracks per cm$^2$ per pCi/l. hr for thoron; (ii) for similar levels TLD (CaF$_2$(Dy)) chips recorded an equivalent of 1.35 ± 0.16 mR for radon and 0.30 ± 0.09 mR for thoron. Taking advantage of the differential response of the two chambers (one covered with a layer of W-1 filter paper and the other with a 75 mm polyurethane foam), simultaneous measurement of radon and thoron could be achieved.

INTRODUCTION

The decay products of radon and thoron are known to carry positive charge immediately after their formation or after further decay by virtue of $\alpha$ or $\beta$ emissions (Du81). This property has been used by several investigators for collecting the decay products on a metal electrode maintained at high negative potential. We reported that an electret (an electrical analogue of a permanent magnet) can be used in the place of high potential for such collection (Ko81). Teflon electrets (6 cm dia. and 0.08 cm in thickness) used for collection produced an electrostatic field equivalent to that produced by a battery of 1000–3000 V. Our earlier work describes a method to calculate the concentration of radon and thoron from the $\alpha$ counting of decay products collected on electrets (Ko81).

Our object here was to adopt the electret chamber technique for collecting the decay products directly on the surface of a TLD or solid-state nuclear-track detector (SSNTD). Such a procedure allowed in situ registration of radioactivity and was, therefore, useful in determining the long-term average concentrations.

MATERIALS AND METHODS

Electret

Teflon electrets were made in the laboratory either by thermoprocesses (Ko81) or by liquid contact procedures (Ch76). The electrets were characterised by measuring the surface charge densities ranging between $10^{-1}$ and $10^{-4}$ Coulomb/cm$^2$ were used for collecting the decay products. The Teflon electrets had the following dimensions: (i) dia., 6 cm; (ii) charged surface dia., 5 cm; (iii) thickness, 0.08 cm.

*Some of the results contained in this paper were presented at 2nd Special Symp. on Natural Radiation Environment, held in Bombay, Jan. 1981.
A stainless-steel sheet (38 mm dia. and 0.8 mm thick) was fixed to the top centre of the electret (Fig. 1A) with the help of a small piece of double-sided adhesive tape. Then the metal surface close to the electret had an induced positive charge and a compensating negative charge appeared on the top surface of the metal sheet. The charge density on the top surface of the sheet was found to be about the same as that on the surface of the electret. The compensating charge (being not bound) was liable to be discharged even by slight contact with any conducting surface. Therefore, the metal sheet must always be handled with a Teflon tweezer to avoid discharge of the surface potential. If the compensating charge leaked away by accidental contact, it was only necessary to remove the metal sheet, ground it and put it back on to the surface of the electret. This method of using the charge of the electret without directly using the electret had some advantages. The electret remained intact and could be reused. Any non-uniform charge distribution on the surface of the electret was not reflected on the charge density distribution on the surface of the metal sheet.

Figure 1(b) shows the procedure for using TLD on the surface of the metal sheet. Usually two TLD chips mounted on 3 mm high aluminum disc, were positioned in the middle of the metal disc. The compensating charges appeared over TLD chips for collecting the decay products. Figure 1(C) shows the arrangement used for experiments with SSNFDs. Such detectors were covered with a thin layer of aluminized mylar with the conducting surface facing out.

Electret holder and electret chamber

Figure 2 shows the arrangement suitable for holding the electret. The brass plunger, screwed into the holder, held the electret tight. Usually 2 thin aluminum foils were used between the plunger and the electret to ensure uniform contact.

Figure 3 shows the electret chamber and the arrangement for fixing the electret holder at the bottom of the chamber. This chamber was similar to the one used in our earlier work (Ko81) and had a volume of 101. A layer of Whatman No. 1 filter paper covered the 20 mesh wire screen on all the sides except top and bottom. The aluminum cover of the chamber had an opening through which the electret holder could be introduced. This opening could be closed with a metal disc. The bottom aluminum plate had an opening where the electret holder could be held. The electret chamber could be suspended by hooks or left on a surface.
Detector

We used commercially available SSNTD (CR-39) (CaF₂), known for its low background. These detectors were cut into rectangular shapes (15 × 14 × 1.6 mm) and were covered on all sides with a thin aluminized mylar (thickness 1.0 mg/cm²) with the conducting face pointing outwards. Such cover attenuates very little of the α energy. An auxiliary experiment was done to determine the track registration efficiency of these detectors for the decay products of thoron present on the conducting surface of the aluminized mylar. The decay products of thoron were collected on the surface of a thin mylar foil using electrical methods (Ko81) and allowed decay of approximately 10 hr to ensure that the decay products were decaying with a half-life of Th-B. A square piece (10 × 10 mm) was cut out of this sample and counted using a well-calibrated α scintillation counter. The sample side was put on the top of the aluminized mylar covering the SSNTD and a small weight was put on the top to ensure intimate contact. The exposure was continued for 24 hr. The expected integrated α disintegrations during this interval could be easily calculated using the data on (i) initial α disintegration rate (ii) the half-life of Th-B and (iii) the exposure time. Using this procedure the registration efficiency of these detectors was found to be 40 ± 2%. We followed the standard etching procedure for α track development (4.5 hr at 60°C in 6 N KOH) and the subsequent microscopic track counting.

TLD chips of CaF₂(Dy) are known to have high response for α radiation (Ra79). These chips were prepared in our laboratory by fixing the locally prepared TLD material on Kanthal chip (13 × 10 × 0.2 mm) using standard techniques. Such detectors were equivalent to TLD-200 phosphor from Harvey. Two such chips were used at a time for collection of the decay products. Aluminized mylar was not used to cover the TLD, because direct collection of decay products onto the TLD gave nearly 30% more response compared to the response of the TLD when collected on a thin aluminized mylar covering the TLD.

**EXPERIMENTS AND RESULTS**

Controlled generation of radon and thoron was done using the arrangement (Fig. 4) used in our earlier work (Ko81). Air from a radium needle was mixed with the humidity conditioned air (HCA) and injected into an inverted half-cut carboy. The top of the cut carboy was covered with a 25 mm thick polyurethene foam. Air containing radon escaped finally to outside through a perforated cover on the drum. Electret chamber was hung at the centre. Humidity and temperature could be measured continuously. For experiments with thoron a chamber containing 6-10 g of thorium hydroxide packed in double layer of filter was used in place of the chamber containing leaky radium needle.

Before starting the experiment, the electret chamber was hung inside the drum (Fig. 4). The electret holder (Fig 1B, C) complete with detectors (two TLD chips or one SSNTD piece), was introduced into the electret chamber and the top of the chamber closed shut. A small double-filter unit (IAEA76) was introduced with its sampling...
point midway between top and bottom of the electret chamber. This double-filter unit sampled at a flow rate of 5 l/min continuously throughout the period of exposure of the detector. The total flow through the drum was 70 l/min. The relative humidity and the temperature during the experiments were 30% and 25°C, respectively.

At the end of sampling period, the electret holder was removed and checked for the status of charge. The final charge should not be below $10^{-7}$ Coulombs/cm$^2$ for satisfactory sampling (Ko81). The detector samples were allowed a delay of at least three hours in the case of radon experiments (3 days for thoron experiments) before reading TLD or before etching the SSNTD. The double-filter sample was analysed by the standard procedure (IAEA76) to calculate the radon and thoron concentration.

Table 1 gives the results on the response of the track detectors for radon. We see that for a given cumulative concentration pCi/l. hr, the response is independent of the actual

<table>
<thead>
<tr>
<th>Table 1</th>
<th>Response of SSNTD (CR-39) for radon in electret chamber</th>
</tr>
</thead>
<tbody>
<tr>
<td>Exp. No.</td>
<td>Concentration pCi/l. hr (%)</td>
</tr>
<tr>
<td>-----------</td>
<td>-----------------------------</td>
</tr>
<tr>
<td>1</td>
<td>190</td>
</tr>
<tr>
<td>2</td>
<td>191</td>
</tr>
<tr>
<td>3</td>
<td>192</td>
</tr>
<tr>
<td>4</td>
<td>116</td>
</tr>
<tr>
<td>5</td>
<td>317</td>
</tr>
<tr>
<td>6</td>
<td>642</td>
</tr>
<tr>
<td>7</td>
<td>798</td>
</tr>
<tr>
<td>8</td>
<td>105</td>
</tr>
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<td>9</td>
<td>124</td>
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<td>10</td>
<td>154</td>
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<td>11</td>
<td>136</td>
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<td>13</td>
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<td>15</td>
<td>222</td>
</tr>
<tr>
<td>16</td>
<td>157</td>
</tr>
<tr>
<td>17</td>
<td>159</td>
</tr>
<tr>
<td>18</td>
<td>177</td>
</tr>
</tbody>
</table>

Grand mean and standard deviation of $T$/C = 92.36 ± 13.26

Note: Track detectors were etched after a minimum delay of three hours after sampling. Relative humidity was 30% and the temperature was 25°C.
concentration or the time of exposure. This follows because of our procedures of allowing the delay of 3 hr for radon. Table 2 gives a summary of the results for radon and thoron.

An auxiliary experiment was conducted to determine the ratio of response for thoron for chambers lined with W-1 filter (Fig. 3) and 75 mm foam (Fig. 5). Both the chambers were exposed simultaneously for the same concentration of thoron and the ratio of their responses was found to be 10±2. This change in response was due to the associated decay of thoron during the transit through an extra thickness of 75 mm foam. No such change was expected for radon due to the long half-life compared to the transit time (Ko81). The differential response characteristics can be used for simultaneous measurement of radon and thoron.

**DISCUSSION AND CONCLUSION**

The choice of 3 hr of delay before subjecting the sample for analysis in the case of radon was to allow the registration of all the possible post-collection α emissions from the decay products collected on the surface of TLD or SSNTD. By that time the decay products had completely decayed because of their short half-lives. Therefore, the calibra-

**Table 2: Response of SSNTD (CR-99) and TLD (CaF$_2$(Dy)) for radon and thoron in electret chamber**

<table>
<thead>
<tr>
<th>Gas</th>
<th>No. of Days</th>
<th>Mean response per pCi/l/hr</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radon</td>
<td>18</td>
<td>92.36±13.26 tracks/cm$^2$</td>
</tr>
<tr>
<td>Thoron</td>
<td>12</td>
<td>9.05±1.52 tracks/cm$^2$</td>
</tr>
<tr>
<td>Radon</td>
<td>12</td>
<td>1.35±0.16 aB</td>
</tr>
<tr>
<td>Thoron</td>
<td>11</td>
<td>0.30±0.009 aB</td>
</tr>
</tbody>
</table>

*Notes: Detectors were read/recorded after a minimum delay of three hours for radon and three days for thoron. Relative humidity was 30% and temperature was 25°C.*
because of a significantly lower concentration of thoron inside the chamber compared to that in the sampling atmosphere due to the short half-life of thoron.

It is important to remember that the sensitivity of this method depends to some extent on the relative humidity. The corrections for the relative humidity could be applied by using the curve given in our earlier work (Ko81).

Electrets also collect ions produced by ionization due to external radiation causing the lowering of the strength of the electric field. However, this does not pose problems in the normal background levels of 50-100 μR/hr for sampling period up to 3 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 on to the electret. Same detectors can be placed on the metal sheet to continue sampling.

The response of TLD or SSNTD reported in our work is for a given configuration used in these experiments. If the sizes of TLD or SSNTD or the configuration is changed, response characteristics for that configuration have to be determined before using this procedure for actual measurement. There is also a scope for improving the configuration to obtain better response.

Cowper and Davenport (Co79) use aluminized mylar sheet with a high negative potential maintained by a set of batteries for collection of decay products produced inside a chamber similar to the one used in our work. A TLD disc placed under the aluminized mylar records the ionization due to the decay products collected on the aluminized mylar. They use CaF₂(Dy) TLD (Harshaw TLD-200) and get a response of 0.33 mR per pCi/l hr. Higher response in our case was because (i) the collection of decay products was directly on TLD material and (ii) the TLDs used in our case had larger surface area.

SSNTD (CR-39) have been used earlier for recording the α directly from an atmosphere containing radon (Ca78). Note that in this case there is no collection of decay production to SSNTD. Therefore, the sensitive volume from which α can register their tracks is very small. The reported value of 0.1264 tracks/cm² per pCi/l hr for radon in a SSNTD similar to CR-39 (Fi80) can be compared to 92 tracks/cm² per pCi/l hr obtained in our experiments. Sensitivity improvement by a factor of 728 is achieved in the present system.

REFERENCES


Ch76 Chudleigh P. W., 1976, “Mechanism of Charge Transfer to a Polymer Surface by a Conducting Liquid Contact”, J. Appl. Phys. 47, 4475.


Makrofol film squares were cut to a standard size of 1' x 1' in order to fit conveniently into sample dan control carriers of the spectrophotometer.

**ABSTRACT**

Dosimetric studies were carried out using Makrofol polycarbonate plastic (195 µm thick) as the radiation sensitive material. It was found that it could be utilized in measuring absorbed doses in the range of $10^6$ to $10^7$ rads using optical density measurement method and in the range of $10^7$ to $2.5 \times 10^8$ rads using chemical etch rate measurement method.

**INTRODUCTION**

The various fields, where application of Solid State Track Detectors (SSTD) have played an important role, also include the branch of radiation dosimetry. This is because SSTD fulfill the basic requirements of a dosimeter such as low cost, simplicity and high degree of reproducibility.

Day and Stein have suggested the use of changes in optical density of plastics, due to irradiation (absorbed dose) as a means of dosimetry. Such changes in optical density have been observed for polycarbonate films due to gamma irradiation and should serve the purpose of dosimetry.

Bentorri has suggested the possibility of using track detecting plastics as high exposure gamma ray detectors based on the dependence of chemical etch rate on absorbed dose and has shown that cellulose nitrate plastic can be utilized in the range of about $10^6$ to $10^7$ rads. Frank and Benton have investigated Lexan polycarbonate plastic and have found that this can be utilized in measuring absorbed doses in the range of about $10^7$ to $10^8$ rads.

In the present work, dosimetric characteristics of Makrofol N (195 µm thick) film have been studied for gamma radiation, using both optical density and bulk etch rate measurement methods. These methods were found to supplement each other and thus enhance the range over which dosimeter could be employed.

**EXPERIMENTAL**

The plastic pieces used in the investigation were successively washed with detergent, tap water and distilled water, dried on filter paper and subsequently handled with tweezers. Irradiation of plastic pieces to different doses was carried out in a $^{60}$Co gamma chamber having a dose rate of $1.2 \times 10^5$ rads per hour.

(A) **Radiation Dosimetry by Optical Density Method**

For the measurement of optical densities, Cary Recording Spectrophotometer model-14 was used. This is capable of measuring optical densities in the range of 0.01 to 2.00.

![Wavelength in nm](image)

![Optical Density](image)

*Fig. 1. Radiation induced ultraviolet absorption in makrofol (195 µm thick).*

Figure 1 shows the absorption spectrum of an irradiated Makrofol piece against an unirradiated piece as reference. A clear peak is found to exist at about
284 nm. This peak optical density value was found to increase with absorbed dose. All the subsequent measurements of optical density were carried out at the wavelengths corresponding to the peak region. This not only gives the maximum value for a given 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 rays. These species differ from the bulk polymer in that they possess electronic configurations that strongly absorb the UV-radiation at that wavelength.

The background optical densities of the plastic films (195 μm thick) to be irradiated were measured at wavelengths corresponding to the peak region, i.e., around 284 nm. They were then irradiated to different doses in a Co^{60} gamma chamber and their optical densities were measured at the same wavelengths. The increase in optical density is a measure of the induced absorption. The dose range covered was from 0-6 x 10^{6} to 1-08 x 10^{7} rads.

When the maximum optical density values were plotted against the corresponding absorbed doses, a linear relationship was found to exist, which can be expressed by the equation \( y = 0.1714x \) where \( y = \text{O.D. of the irradiated piece} \) and \( x = \text{Absorbed dose in Megarads} \). The background optical densities of the plastic films were measured at the peak region. No fading in optical density values was noticed even after a period of two months. By this method, it is convenient to carry out number of dose measurements within a short time.

In the present investigation, Makrofol plastic of 195 μm thickness was used. If it is desired to measure smaller radiation doses, measurable optical density can be obtained by using thick films. When large doses are to be measured, one can employ thinner films, e.g., 40 μm thick film can cover the range up to about 4-0 x 10^{7} rads.

(B) Radiation Dosimetry by Etch Rate Method

Makrofol film circles of about 1" dia. were individually weighed on a balance capable of reading thousandth of a mg. The plastic circles were exposed to Co^{60} gamma rays in a gamma chamber in the dose range of 10^{6} to 2-5 x 10^{6} rads. At high doses in the range studied, the plastic films were found to become sticky. Therefore aluminium foils were used as spacers between the plastic circles during irradiation. Above this dose range, Makrofol plastic was found to become brittle and difficult to handle. Hence the study could not be extended to higher doses.

The plastic circles were weighed, after different doses of gamma irradiation but before etching, to check whether there was any weight loss because of irradiation. It was observed that at doses of 5-0 x 10^{6} rads and above there were measurable weight losses which are assumed to be due to escaping gases.

Four Makrofol circles were irradiated for each gamma dose. In the present investigation etchings were carried out at 80°C in 6N NaOH. Eleven circles comprising one from each irradiation and one control were etched simultaneously in an etching set-up in which each circle was held in a small separate compartment formed by interconnecting perspex rings and stainless steel wiremesh. The weight losses due to etching were measured for etching periods of 1, 2, 3, and 3-5 hours. From these values, the thickness losses from a single surface of films were calculated. When the average thickness losses (for four films) were plotted against various etching periods of each dose, straight lines were obtained. This indicated that the etch rate remained constant for various etching periods, i.e., the etch rate did not change with depth in the film.

Similarly when the average thickness losses from a single surface for various etching periods were plotted against the corresponding absorbed doses on a semi-log paper, parallel straight lines were obtained. This, therefore, indicates that the average thickness loss of an irradiated film is an exponential function of the absorbed dose. These parallel straight lines (on semi-log paper) are represented by the general equation \( y = 0.00246x + C \), where \( y = \text{Average thickness loss in μm from a single surface (on log scale)} \), \( x = \text{Absorbed dose in Megarads (linear scale)} \) and \( C \) has values of 0-524, 0-808, 0-960 and 1-028 for etching periods of 1, 2, 3 and 3-5 hrs respectively.

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The technique which led to the development of track-etched, micro-filters was discovered in 1962 by Price P. B. and Walker R. M. 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 cleavable solids and plastics.

The heavy charged particles passing through any of these solids produce continuous damage along their path and thus leave behind a trail of radiation damaged material. The chemical etching of these irradiated solids leads to the formation of fine hollow channels along the path of the charged particles due to preferential etching of the damage trail. These channels are usually uniform in width along their entire length and maintain the directions of the original tracks. If the thickness of the detector sheet is less than the particle's range in it, the above process leads to the formation of fine holes in the irradiated detector sheet. The micro-filters thus formed have uniform holes and offer certain advantages over conventional filters made of cellulosic plastics and having irregular holes. Fig. 1 is a microphotograph of a micro-filter.

Experimental

The uniformity of the holes is achieved by allowing a collimated beam of charged particles to strike the filter sheet. A simple wav of collimating the beam of particles is 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 will be the source alignment.

An electrodeposited californium-252 source (10 mm diameter) was used as a source of fission fragments. The distance between the source and the filter was 12 cm. Since the diameter of the irradiated circular film portions was 2.5 cm, the angle of incidence of fission fragments remained 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. A one-to-one correspondence exists between fission fragment tracks and the etched holes under conditions approximating to normal incidence.

The hole diameters at different etching times revealed that the diameter increased linearly with the etching time. The largest hole size that could be obtained depended on the film thickness. Thus the holes of 9-6, 10-6 and 14-2 μm diameter were obtained in films of 10, 12 and 16 μm thickness respectively. In these large hole size regions, however, the films became very thin and posed handling problems. The smallest hole size that could be conveniently obtained was ≈ 1 μm.

The hole densities depend directly on the particle flux. But randomness in positions of holes must be considered as a limiting factor. This leads to formation of large sized holes due to overlap, which impair the usefulness of the filter. The maximum hole density obtainable is actually governed by the acceptable probability of overlap and the hole size. In any work involving the use of these filters, it is extremely unlikely that filters with overlap probabilities exceeding 10% could be used. Since the percentage area occupied by these holes is only slightly greater than the percentage probability of overlap (irrespective of hole size) the strength of the filter sheet will not change significantly due to porosity of this order.

Applications

The above microfilters were tested and found useful in the following applications—

(A) Biomedical applications

(i) Separation of lymphocytes from tumor cell suspensions prepared from solid tumor. Fig. 2 is a microphotograph of Murine Fibrosarcoma Tumor Cells separated by using a filter with hole diameter = 2.86 μm.