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Factors affecting the registration and counting of alpha tracks in solid state nuclear track detectors

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Abstract : In view of the fact that the radon progeny contribute the highest to the natural radiation dose to general populations, large scale and long-term measurements of radon and its progeny in the houses have been receiving considerable attention. Solid State Nuclear Track Detector (SSNTD) based systems, being the best suited for large scale passive monitoring, have been widely used for the radon gas (using a cup closed with a semi-permeable membrane) and to a limited extent, for the measurement of radon progeny (using bare mode in conjunction with the cup). These have been employed for radon mapping and indoor radon epidemiological studies with good results. In this technique, alpha tracks recorded on SSNTD films are converted to radon/thoron concentrations using corresponding conversion factors obtained from calibration experiments carried out in controlled environments.

The detector response to alpha particles depends mainly on the registration efficiency of the alpha tracks on the detector films and the subsequent counting efficiency. While the former depends on the exposure design, the latter depends on the protocols followed for developing and counting of the tracks. The paper discusses on parameters like etchant temperature, stirring of the etchant and duration of etching and their influence on the etching rates on LR-115 films. Concept of break down thickness of the SSNTD film in spark counting technique is discussed with experimental results. Error estimates on measurement results as a function of background tracks of the films are also discussed in the paper.

Keywords : Alpha tracks, radon progeny, SSNTD.

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1. Introduction

The science of solid state nuclear track detectors was born in 1985 when D A Young discovered the first tracks in crystal of LiF [1]. Operation of the solid-state nuclear track detector is based on the fact that a heavy charged particle will cause extensive ionization of the material when it passes through a medium. For example, an alpha particle with energy of 6 MeV creates about 150,000 of ion pairs in cellulose nitrates. Along the path of the alpha particle, a zone enriched with free chemical radicals and other chemical species is then created. This damage zone is called a latent track. The

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track effect exists in many materials. It is particularly pronounced in materials with long molecules, *e.g.*, cellulose nitrates or different polycarbonates, and such materials are the most convenient ones for application and detector manufacturing. A comprehensive survey of the materials that show the track effect is given by Fleischer *et al* [2]. Now when a piece of material containing latent tracks is exposed to some chemically aggressive solution, chemical reaction would be more intensive along the latent track. The overall effect is that the chemical solution etches the surface of the detector material, but with a faster rate in the damaged region. In this way a 'track of the particle is formed, which may be seen under an optical microscope or by other counting techniques.

2. Track development in materials

One of the challenges that have attracted significant amounts of attention was a formal description of the track development *i.e.*, growth of tracks. The problem is rather geometrical in nature. In addition, there are theories that describe the physical aspect of track formation. However, until now, there is not a single complete theory that satisfactorily explains track formation and calculates the parameters related to the tracks. The simplest case of track development refers to that when the incident particle enters a detector under normal incidence with respect to the detector surface (Figure 1). In this I is the initial detector surface, I' is the surface after etching, V_t is the etch rate along the particle trajectory (track etch rate) (taken constant here), V_b is etch rate under the undamaged region of the detector (bulk etch rate), O is the entry point of the alpha particle and E is the end point of the particle in the detector material. OE = R is the particle range in the detector material. The distance I and I' is equal to h, *i.e.*, the thickness of the later removed by etching, L' is the total distance traveled by the etching solution along the particle track, and L is the track length.

In one aspect the track development is analogous to wave propagation. In the case of the development, a hemisphere with a radius $h = V_b t$ (t is the etching time) is formed around each point on the detector surface, except in the direction of the particle path where the etching progress with the rate V_t. Even though the track formation is a three dimensional phenomenon, for the sake of simplicity, the track development can be illustrated in two dimensions as shown in Figure 1.

The track length 'L' is given by :

$$L = \left(V_t - V_b\right) t \,. \tag{1}$$

From the figure we can see that :

$$\tan\theta = \frac{D/2}{L} \,. \tag{2}$$

From the similarity of triangle in Figure 1 it can be observed that :

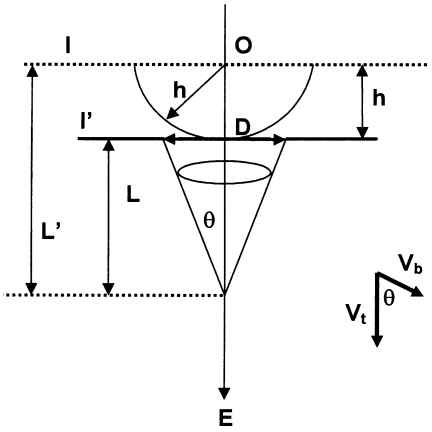

Figure 1. Geometry of the track development.

$$\sin\theta = \frac{V_b}{V_t}\,. \tag{3}$$

Combining eqs. (1), (2) and (3) it can be shown that :

$$D = 2V_b t\sqrt{\frac{V_t - V_b}{V_t + V_b}}\,. \tag{4}$$

If $V_t >> 1$, eq. (4) is reduced to :

$$D = 2V_b t = 2h\,. \tag{5}$$

Based on eq. (5) an indirect method for bulk etch rate measurements can be developed. However, this is applicable for heavy ion or fission products used for irradiation. Similar models are developed for oblique incidences of charged particles. The above models are derived based on constant track etch velocity (V_t). However, V_t, is not constant in most of the realistic cases. It could vary along the path of the track and can be maximum at the end of the track. However, a realistic approach is to consider the velocity of track etch maximum just before end of the track. Three cases of track etch velocities are represented in Figure 2.

Since track etch rate V_t, is an inbuilt function in etching, we consider the prime factor of bulk etch rate V_b as one of the most important parameters that control the formation and development of tracks. There has been a large volume of literature discussing V_b. It has been shown that V_b depends on many factors like the purity of the basic substance, the molecular structure of the polymers, conditions of polymerization, environmental conditions during the irradiation and finally on etching conditions [3]. In this way there are different topic related to V_b which include the following :

1. dependence of V_b on the chemical composition and preparation of the detector;
2. dependence of V_b on the etching conditions (temperature and concentration of etchant) as well as stirring conditions;

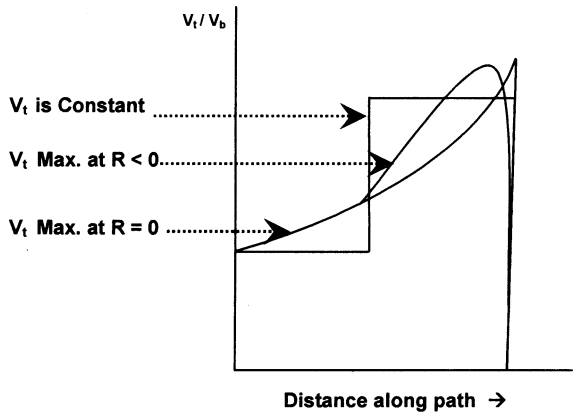

Figure 2. Variation of the V_t function along the particle path.

3. dependence of V_b on the irradiation of the detector before etching with different kinds of ionizing or non-ionizing radiations.

3. Bulk etch rate (V_b)

The bulk etch rate is the rate of removing of the undamaged surface of the detector. Due to chemical reaction between the etching solution (etchant) and the detector material, some molecules of the detector are removed. The final effect is the removal of the material from the detector surface. During etching, the material is removed layer by layer and the thickness of the detector becomes smaller and smaller.

3.1. Dependence of etching conditions on bulk etch rate :

Studies have been carried out by various workers on different etching conditions like etchant molarities, duration of etching and temperature of the etchant for the different type of detectors used. It has been established that the etching of plastic detectors increases exponentially with temperature of the etchant as shown in eq. (6).

$$V = V_0 e^{-\varepsilon_0/kT} \tag{6}$$

where T is the temperature in K, ε_0 is activation energy in eV, k is the Boltzmann's constant and V_0 is a proportionality constant. Eq. (6) is applicable to both V_b and V_t with different activation energies. A typical example of bulk etch rate for CR-39 films with NaOH is shown in Figure 3 [4].

Results clearly indicates that the bulk etch rate is following an exponential growth with increase in molarity of the etchant. Gruhn *et al* [4] have also studied the etching characteristics of cellulose nitrate plastic detectors as a function to temperature and normality of the etching solution. The normality of the etching solution was varied between 2.5 and 7.69 N and the temperature was varied between 25 and 55°C. The characteristics of V_b growth with the normality depended on the temperature. At 25°C, the growth had represented as exponential but at higher temperature the growth was

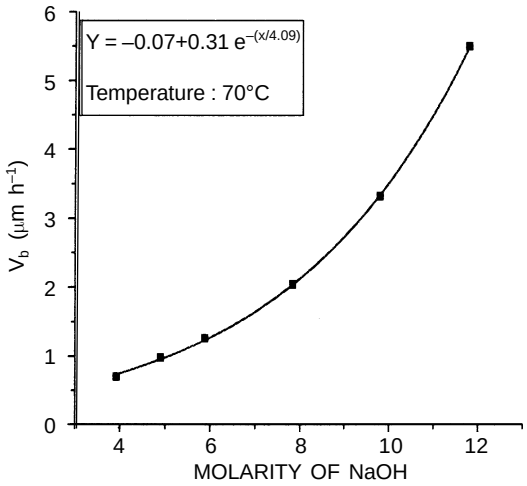

Figure 3. Bulk etch rate with molarity of etchant [4].

not exponential and there was a saturation effect for larger normality. Enge *et al* (1974) also have shown in their studies on bulk etch rate for cellulose nitrate films, that a saturation on V_b is attained beyond 3 N NaOH solution.

3.2. Effect of stirring on bulk etch rate :

Enge *et al* [5] hypothesized that a colloid layer of partially dissolved cellulose nitrate molecules together with components of the etching solutions adhered to the surface of the plastic during the etching process. They studied the effects of stirring speed on cellulose nitrate for NaOH at 50°C and found that the bulk etch rate increased with the stirring speed for a constant normality of NaOH, which could be explained by washing off the colloid layer. Yip *et al* [6] have reported from their studies the differences in bulk etch rates for stirring and non-stirring conditions. In their experiment they used 10% aqueous solution of NaOH maintained at 60°C for etching. They reported the bulk etching rate under magnetic stirring as 6.65 ± 0.34 μm h^{-1} and that under no stirring as 3.61 ± 0.14 μm h^{-1}.

3.3. Influence of detector ageing on bulk etch rate :

Studies have shown that age of the detector can also affect the bulk etch rate. Siems *et al* [7] have studied the etching behavior of cellulose nitrate films (LR-115) manufactured at different years. They observed that (i) the bulk etch rate of the detectors of the same batch is constant over 5 years, (ii) detectors of different batches older than 5 years that were stored at +20°C show an odd bulk etching rate behavior and (iii) the difference in bulk rate with age can be reduced if the films are stored at +4°C.

3.4. Influence of irradiation on bulk etch rate :

Studies have shown that the plastic detectors irradiated with gamma rays, electors,

protons, ultraviolet and infrared radiation have substantial effects on its etching character.

The three parameters of detector, V_b, V_t and V increase by increasing gamma absorbed doses, except that a clear drop is observed in the bulk etch rate at low gamma doses. The effect of gamma irradiation is highly pronounced on the detector surface and decreases with depth from the surface. Thus V_b decrease with etching time.

4. Track etch rate (V_t)

The track etch rate is the rate of detector etching along the particle track, which is along the preferential direction of etching. A track is formed when $V_t > V_b$. For an oblique incidence of the particle, a track is formed when $\sin \theta > V_b/V_t$.

Jonsson [8] and Somogyi [9] proposed that the velocity of etching at a point in the etchable portion of the track depends on the distance of that point from point of maximum damage along the track. The latter point occurs slightly before the point where the alpha particle stops, or would have stopped, in the material. The empirical equation describing the velocity of etching along the track (V_T) is :

$$V_T(R) = V_B \left\{1 + e^{(b-aR)}\right\} \tag{7}$$

where R is the arbitrary distance reckoned from the end of the track, b determines theoretically the velocity of etching at the stopping end of the particle track and a determines the decrease in velocity per incremental increase in the distance from the stopping point. The constants a and b are characteristics of the detector. Andriamanantena and Enge [10] from their experiments have suggested the values 0.16 μm^{-1} for a and 2.68 for b for etching with 2.5 N NaOH etchant at 60°C.

5. Alpha track counting

Etched tracks are counted by optical method, but the measurement technique by optical microscope is time consuming and tedious. The spark counting of chemically etched charged particle tracks in organic polymers (SSNTDs) is a well documented technique [11]. Usually an aluminized film is used as the electrical conductor between the spark and return electrodes. The heat of the spark evaporates the aluminum over a small area, thereby electrically isolating once-sparked holes. Double counting is therefore minimized if not entirely eliminated.

Some of the etched tracks are sufficiently close for one, or occasionally more, of them to be permanently masked by the area of spark induced electrical isolation surrounding that hole which is sparked first amongst such pairs or groups in each sequence. Obviously this effect, which results in non-linearity, gradually increases as the track density increases.

In radon thoron measurements using SSNTDs, the alphas striking the detector material being sparked off from different distances with respect to detector surface, the track lengths formed inside the film are also different. Some of the holes which are partially opened by etching may not have sufficient material thickness to withstand the voltage applied resulting in to create additional holes in the film. This thickness is called the break down thickness for spark counter which is unique for each spark counter. Eappen and Mayya [12] in their studies had found that the breakdown thickness is 3 μm for the system they used.

Two uncertainties are thus observed in spark counting, (i) error due to breakdown thickness, which can be avoided when operating voltages for the spark counter are kept constant. Additional holes, compared to optical counting, created due to break down thickness of the material will be absorbed in the calibration factor obtained using spark counting. (ii) The error due to overlapping of the spark area can be corrected using eq. (8).

$$N_T = N_S\left[1+\frac{a\times N_S}{2}\right] \tag{8}$$

$a = \pi(r_1 - r_2)^2$; r_1 is the radius of the Al foil hole > 100 μm

r_2 is the radius of the track hole $\cong$ 6 μm

where N_T is the actual number of alpha tracks developed and N_S is the number of tracks counted using spark counter.

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