

## ANOMALOUS BEHAVIOUR OF ENVIRONMENT AFFECTED CR-39 AT ELEVATED TEMPERATURES

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The evidence obtained indicates that the stability of latent tracks in CR-39 is affected by the environmental conditions which the detector experiences during annealing. The depth dependence bulk etch rate, balloon-like surface features are some of the anomalous observations that are discussed in the text. It has been observed that the ballooning affect is a function of annealing time and temperature. An attempt has been made to explain the chemical changes accompanying the anomalous behaviour of the detector.

### 1. Introduction

In conceiving new experiments involving charge identification one needs to know the factors that limit detector resolution. Track registration by organic polymers is affected by a large variety of physical, or chemical treatments of the detector material [1-3]. These include manufacturing processes, environmental conditions, ageing, thermal treatments, ultraviolet and  $\gamma$ -ray exposure. In addition, oxidation, photolysis, thermal degradation, plasticizer volatilization, photosensitization and crystallization are some of the phenomena responsible for alteration of organic polymers and the track detector response. It has been further observed that the solubility of the polymers increased with gamma dose [4]. Crawford et al. [5] discussed the observation that ultraviolet light in the presence of oxygen can greatly increase the etching rate of tracks in Lexan without significantly changing the bulk etch rate,  $V_b$ . Shah et al. [6] later supported these findings in Makrofol. The affect of active reagents after manufacture has been intensively studied. Portwood et al. [7] found that the sensitivity  $V_t/V_b$  of CR-39 tends to decrease within three weeks of manufacture. This fading continues at a much reduced rate thereafter. It has been shown that it is the plastic that fades not the latent track. Similar effects have been observed in block copolymers styrene-butadiene-styrene [8]. Treatments during irradiation with active agents such as  $O_2$ ,  $O_3$ ,  $H_2O_2$  [9] and even  $H_2O$  have been observed to increase the track etching rate of polymers, whereas natural environments such as vacuum or an  $N_2$  atmosphere decrease  $V_t$ , presumably by excluding oxygen. It is felt that these active species tend to stick to the open ends of the broken chains, thereby preventing broken polymer chains from reuniting and repairing the damage.

Two fundamentally different mechanisms affect the properties of organic molecules under heating. The first mechanism is a reversible process. By heating or cooling a polymer material, it is possible to change its morphology in different ways, dependent on whether the organic polymer is crystalline or wholly amorphous. Crystalline polymers may become amorphous if quickly quenched from their molten state below a certain temperature called the glass transition temperature,  $T_g$ .

The second mechanism, the one about which we are more concerned, is the change that takes place in plastics due to the heat absorbed by the polymer after irradiation. Investigations have been carried out [10,11] on the effects of thermal treatment in air on the registration characteristics of different organic detectors. It has been observed in polycarbonates that the efficiency as a function of annealing temperature shows a rapid drop just around the glass transition temperature. Different views have been presented on the etch rate studies carried out on CR-39 in an oxygen/air atmosphere after annealing. Heins and Enge [12] have observed that the normalized track etch rate,  $V_t/V_b$ , remains almost constant. Annealing the plastic in hot air at 98°C, Benton et al. [13] on the contrary support an increase of the  $V_t/V_b$  ratio. Henshaw et al. [14] observed that the track etch rate  $V_t$  decreased whereas  $V_b$ , the bulk etch rate increased, resulting in an overall decrease in the ratio  $V_t/V_b$ . In their paper, Adams and Beahm [15] examined the long term annealing of stopping Fe tracks in Lexan and found as much as a 20% reduction in the etching velocity at 40°C, whereas no such effect was observed at temperatures lower than room temperature. Bulk etch rate variations have been observed in Makrofol N [16] and CR-39 [14]. Makrofol E, Lexan and Tuffak [15-17] showed no sign of variations in the bulk etch rates. In addition, certain anomalous etching char-

acteristics of CR-39 have been projected by Henshaw et al. [14].

The diverse behaviour of various plastics have motivated us to carry out an extensive study on the post annealing behaviour of CR-39.

## 2. Experimental details

A survey of the literature as well as experimentation helped us to choose the etchants and the etching temperature for CR-39 [18]. The alkali etchant 6M NaOH at 60°C was found to be ideal for our experiments. The Haake N3 circulator was used to keep the etchant temperature constant, the accuracy of control being  $\pm 1^\circ\text{C}$ . All measurements were made using Carl Zeiss Microscope.

Samples of CR-39 (Pershore Mouldings, UK) were cut from large sheets of the plastics. The samples were sent to GSI, Darmstadt for irradiation with energetic heavy ion beams at incident angles of  $90^\circ$  and  $45^\circ$ . CR-39 was irradiated with  $^{208}\text{Pb}$  (17 MeV/n),  $^{139}\text{La}$  (14.6 MeV/n) and  $^{93}\text{Nb}$  (18 MeV/n). The irradiated CR-39 samples were cut up into small test samples. These samples were cleaned in distilled water and annealed in a furnace with a temperature control accuracy of  $\pm 1^\circ\text{C}$ . The samples exposed to various ion beams were annealed between temperatures of  $160^\circ\text{C}$  to  $240^\circ\text{C}$  for time intervals of 10, 20, 40 and 60 min; care was taken for the annealing of samples of various ions simultaneously.

## 3. The variation of the bulk etch rate under heat treatment

The bulk etch rate  $V_b$  was measured using the track diameter technique ( $V_b = D/2t$ ). For this purpose, we chose a normally incident ion beams. The graphs in figs. 1–4 represent the variation of the etched diameter with etching time under different combinations of annealing

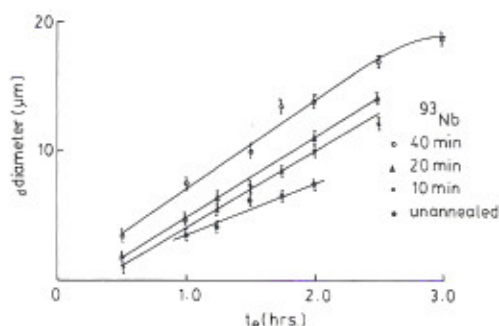


Fig. 1.  $V_b$  measurement of  $^{93}\text{Nb}$  tracks at  $150^\circ\text{C}$  at different annealing times.

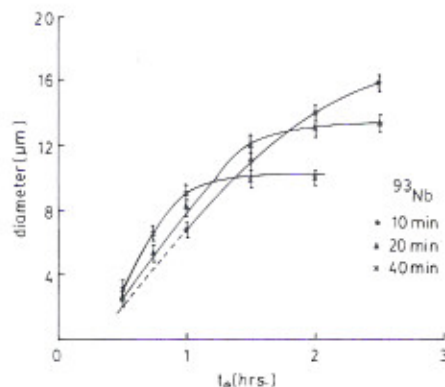


Fig. 2. The measurement of  $V_b$  at  $175^\circ\text{C}$  at different annealing durations.

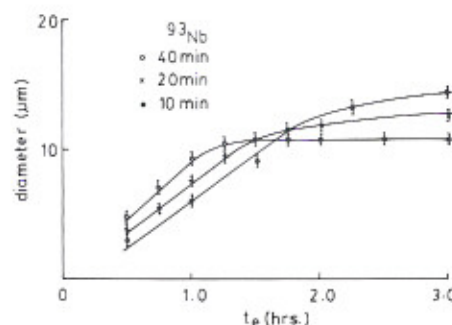


Fig. 3. Plot between diameter and etching time of  $^{93}\text{Nb}$  ion tracks at annealing temperatures  $200^\circ\text{C}$ .

temperature and time for  $^{93}\text{Nb}$  ion in CR-39. The experimental data on  $^{139}\text{La}$  and  $^{208}\text{Pb}$  annealed simultaneously with  $^{93}\text{Nb}$  in an air atmosphere indicates that the bulk etch rate  $V_b$ , indeed varies with the extent of annealing [14,17]. The variation of  $V_b$  from a value of  $1.90 \mu\text{m/h}$  for unannealed  $^{93}\text{Nb}$  ions in CR-39 samples to  $9.79 \mu\text{m/h}$  at an annealing temperature of  $240^\circ\text{C}$  for 60 min indicates the effect of annealing of CR-39 in air. Depth dependent bulk etch rate has been noticed in

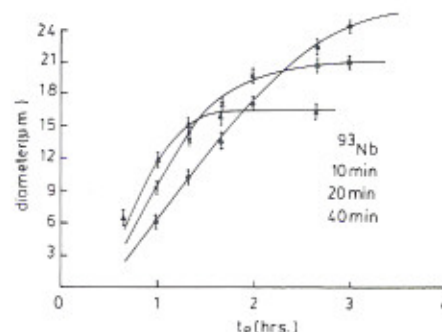


Fig. 4. Bulk etch rate measurement using the diameter method at  $220^\circ\text{C}$  for  $^{93}\text{Nb}$  ion tracks.



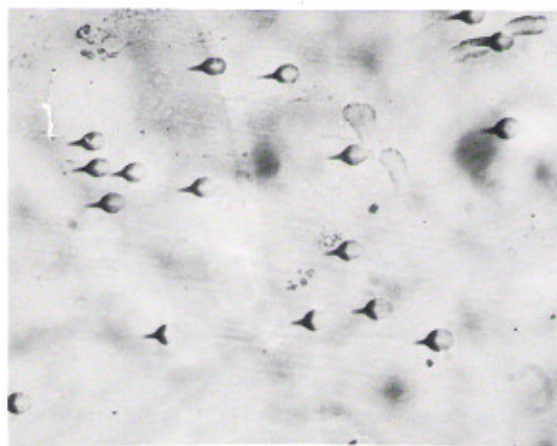


Fig. 5. Microphotograph of  $^{93}\text{Nb}$  ions showing ballooning effect.

our CR-39 samples and it is most marked near the plastic surface. The balloon-like feature is prevalent in all oblique penetrating tracks in CR-39 (fig. 5).

#### 4. Discussion

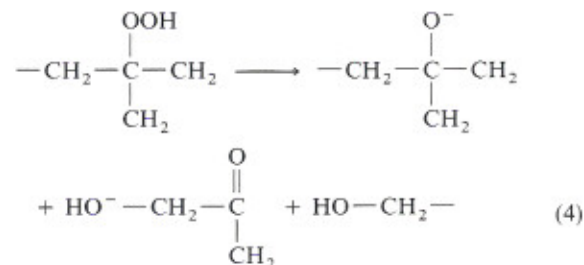
A possible reason for the increase in  $V_b$  in CR-39 can be the oxidation of the CR-39 samples by the atmospheric air. Generally in plastics, free radicals will be formed especially from the decomposition of primarily formed hydroperoxides [7].



The formation of hydroperoxides on the substrate will preferentially occur with tertiary hydrogen atoms. In CR-39 the tertiary hydrogen in the structure will be



susceptible to such attack. The hydroperoxides can then decompose to form ketones, alcohols and oxygen. In the case of CR-39, a possible scheme would be thus leading



to scission of the hydrocarbon chain and hence an increase in the bulk etch rate,  $V_b$ .

From our results it is quite obvious that the temperature of annealing and time of annealing enhances the  $V_b$  rates. The transfer of heat energy to the samples helps in the decomposition of the primarily formed hydroperoxide [eqs. (1) and (4)] and further oxidation of the samples, increasing the percentage scission of the hydrocarbon chain and hence an enhancement in  $V_b$  with the extent of annealing. The bubble-like formation at the surface of the oblique tracks (fig. 5) indicates that it is the surface which is effected by oxidation to a greater extent, as compared to the inner region of the samples.

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