Ageing effects on polymeric track detectors: studies of etched tracks at nanosize scale using atomic force microscope

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Among several different techniques to analyze material surface, the use of Atomic Force Microscope (AFM) is one of the finest method. As we know, the sensitivity to detect energetic ions is extremely affected during the storage time and conditions of the polymeric material used as a nuclear track detector. On the basis of the surface analysis of several track detector materials, we examined the detection sensitivity of these detectors exposed to alpha particles. The preliminary results revealed that the ageing effect on its sensitivity is very strong, that need to be considered on the routine applications or research experiments. The results are consistent with the experimental data in the literature.

Keywords: Ageing; SSNTDs; nanosize scale; alpha-tracks; Atomic force microscope.

Entre las diferentes técnicas para analizar la superficie de los materiales, uno de los mejores métodos es el uso del Microscopio de Fuerza Atómica (AFM). Como sabemos, la sensibilidad para detectar iones energéticos se afecta durante el tiempo de almacenamiento y las condiciones del material polimérico utilizado como Detector de Trazas Nucleares. Con base en el análisis de la superficie de varios materiales, se determinó la sensibilidad de detección de partículas alfa. Los resultados preliminares revelaron que el efecto del envejecimiento de su sensibilidad es muy fuerte, y debe tenerse en cuenta en las aplicaciones rutinarias o de experimentos de investigación. Los resultados son consistentes con los datos experimentales en la literatura.

Descriptores: Envejecimiento; SSNTDs; nanoescala; trazas de partículas alfa; microscopio de fuerza atómica.

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1. Introduction

The high sensitivity and the long-term stability of nuclear track etch detectors are extremely important in various experiments and many applications. Consequently, CR-39 as a unique polymer track detector is now used in wide applications: in nuclear and particle physics, earth sciences, reactor and radiation physics. In addition, it has also been used in engineering experiments that involve routine exposure of CR-39 plates to ionizing radiation and light. Lately, another new polymeric track detector named SR-90 was produced by Fujii and his collaborators [1]. In spite that this detector has higher detection sensitivity than that of CR-39 for heavy ionizing particles in addition to protons, the ageing effect on the sensitivity of SR-90 is stronger than that of CR-39 [1]. The ageing effect on the sensitivity of track detectors in air and various gases such as O_2, O_3, N_2, Ar, H_2O, and NO_2 has been investigated by many research groups [2-9].

All ageing studies mentioned above were based on indirect measurements of track depths at a micrometer scale resulting from long etching periods. These indirect measurements were usually performed using optical techniques. Another line with extreme resolutions was developed based on direct measurements using atomic force microscopy. Track depths at a nanometer scale, resulting from very short etching time (in the early stage of the etching process), can be directly observed using atomic force microscopy.

Atomic force microscope (AFM), as a fine tool at extremely short distance scale, has been used in the study of track sensitivity and surface roughness of SSNTDs [10-12]. Moreover, many related subjects such as latent track formation, etching mechanisms and latent nuclear tracks in organic material had been directly observed using atomic force microscope, respectively [13-15]. In the present work, AFM has been applied to evaluate effects of ageing and storage conditions on track sensitivity and surface roughness for various kinds of polymeric nuclear track detectors.

2. Experimental procedures

The detector samples used in this work were classified into two groups according to their storage conditions and ageing time. The first was stored in freezer at -4 °C and the second

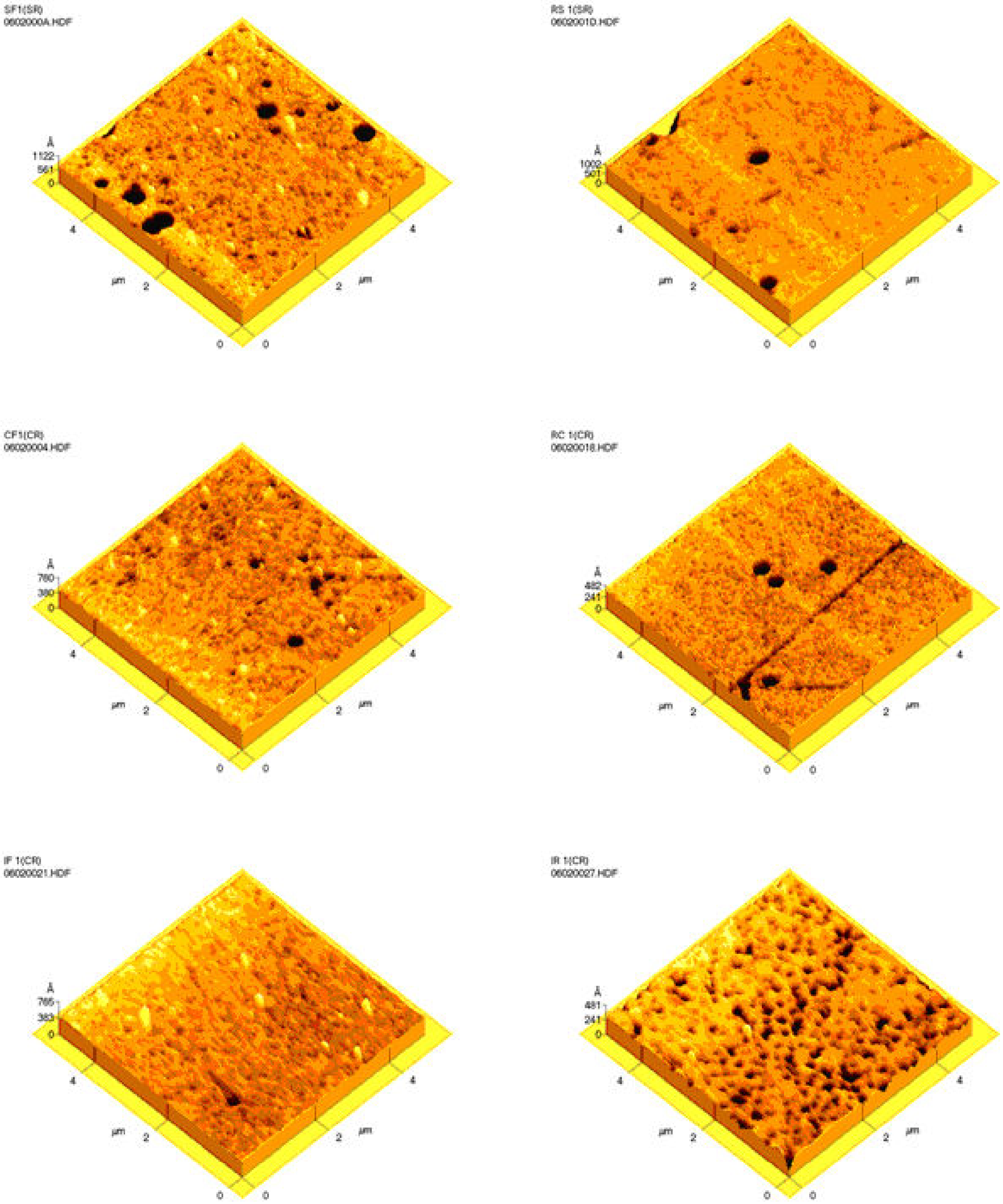

FIGURE 1. 3-D images showing the formed alpha tracks in the plastic detectors SR-90, CR-39 (HM) and CR-39 (IC) stored in freezer (left side top, middle and bottom) and in air at room temperature (right side top, middle and bottom).

was kept in air at room temperature for different periods. These detectors are CR-39 (IC: Intercast) supplied by Intercast Europe S.P.A. of Parma, Italy, CR-39 (HM: Homemade) and SR-90 (HM: Homemade) polymerized in Fujii laboratory at Faculty of Engineering, Aomori University, Japan [1] and sent to our laboratory in Zagazig. The Homemade CR-39 (HM) and SR-90 (HM), in addition to Intercast CR-39 (IC) were aged for two and seven years, respectively. The samples of CR-39 (IC) and both of homemade detectors CR-39 (HM) and SR-90 (HM), 660 and 930 μm thick respectively, were prepared from larger sheets by cutting them into pieces 1 cm $\times$ 2 cm size. After the storage in freezer and in air at room temperature for a proposed time as mentioned before, the samples were irradiated with α-particles with energies 5.0 MeV under normal incidence. The α-source employed in the present work was ^{241}Am source (initial energy of α-particles

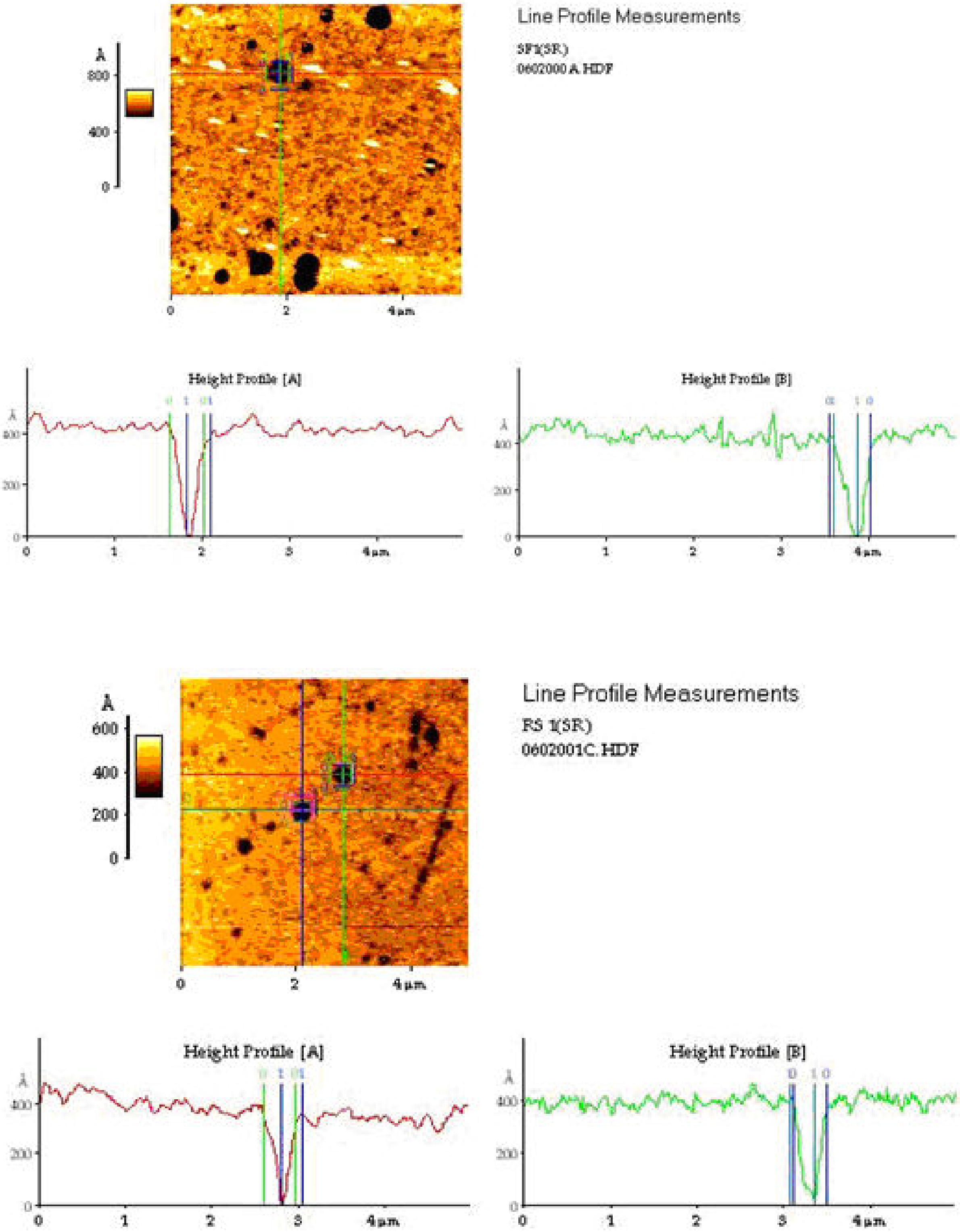

FIGURE 2. 2-D images of the etched tracks in SR-90 with the height profiles of the 5 MeV alpha particles. The detectors aged in freezer (top) and in air at room temperature (bottom).

is 5.49 MeV). After exposure, these detectors samples were etched in 6.25 M KOH solution at 60 ± 1 °C. The etching time was extremely short (5 minutes) and etch pits were at the non-observed stage using any traditional optical method. The surface roughness and the track etch pits were measured using AFM.

The AFM (Digital Instrument CP-II Scanning Probe Microscope-Veeco Metrology Group, USA) equipped with a silicon premounted cantilever having a silicon conical tip 5-7 microns height and 10 nm radius of curvature. The optimized scan parameters were: a force setting of 0.26 N/m, and a raster scan frequency of 1 Hz. The surfaces of the detectors were imaged directly in air, at room temperature.

3. Results and discussion

Fig. 1 shows three-dimensional images of the detector surfaces taken with a scanning area $5 \times 5 \mu\text{m}^2$ of Intercast CR-39 (IC) and both of home-made detectors CR-39 (HM) and SR-90 (HM) which were aged for 7 and 2 years, respectively, at two different storage conditions: in freezer and in air at room temperature. These images show the formed α-tracks at a very short etching time which corresponds to the stage of track birth. Since the alpha particles incident on the detector material had the same energy and incident angle (normal incidence in our study), the tracks should have approximately the same size and depth. From this point of view, one can get accurately a direct imaging of the size and depth of the tracks structure as well as the RMS roughness of surface of aged and non-aged detectors. The surface roughness changed considerably for both kinds of CR-39 during the course of the ageing time in air at room temperature. While the effect of ageing and storage conditions on the surface roughness of SR-90 was not significant, in spite of SR-90 detector showed rougher surface than the both kinds of CR-39 used in this study.

Fig. 2 shows, as an example, two-dimensional AFM images of alpha track etch pits with the surface profiles of SR-90 aged in freezer and in air at room temperature. In both cases it is possible to observe a difference between depths of etch pits. In the case of detectors kept in freezer the track lengths of etch pits are deeper than those of detectors stored in air at room temperature. Generally, a similar trend is observed for homemade CR-39 (HM), while in the case of Intercast CR-39 (IC) there is no any significant change in the track length of the aged samples in freezer than those of samples stored in air at room temperature.

On the basis of the track geometry, the etched track model involves two important etching parameters: the track etch rate V_t (*i.e.* the rate of chemical dissolution along the damaged zone due to ionizing particles) and the bulk etch rate V_b (*i.e.* the etch rate at the undamaged zones of SSNTD). The track etching in SSNTD proceeds at a relatively constant rate in the short period of etching. From the data in Table I which were measured using AFM, it is promising to estimate the track sensitivity S with the minimum uncertainty in terms of etching parameters: L, the visible track length and h, the layer removed during the desired etching time. We can determine the sensitivity S from these geometrical relationships by the following formula:

$$L = V_t t - V_b t \quad (1)$$

where t is the etching time. Therefore,

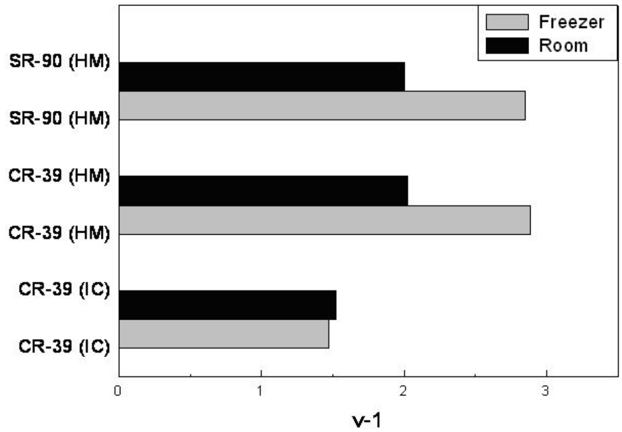

FIGURE 3. Effects of storage conditions on the sensitivity of SR-90, CR-39 (HM) and CR-39 (IC) track detectors. 5 MeV alpha particles from ^{241}Am were irradiated in air.

$$V = V_t/V_b = 1 + L/h \quad (2)$$

$$S = V - 1 = L/h \quad (3)$$

The variations of sensitivities versus the storage time in air at room temperature and in freezer are shown in Fig. 3. It has been reported that the initial sensitivity of SR-90 just after polymerization is about 3 times higher than that of CR-39 [1]. In this figure, the SR-90 sample stored in the freezer shows a small decrease of its sensitivity, while the original sensitivity of SR-90 is a little bit higher than that recorded in our study. This is due to the transportation time from Japan to Egypt, which spent about two weeks in air at room temperature before restoring it once again in the freezer. This decrease in the initial sensitivity of SR-90, if it is stored in air at room temperature for about ten days, is quite consistent with the experimental data measured by one of the authors (M. F.) as reported in their article [1], while the initial high sensitivity of that detector sample which was aged in air at room temperature shows significantly the loss of its original sensitivity that was attained just after polymerization. Also, the same mechanism was approximately observed for home-made CR-39 (HM).

On the other hand, in the case of CR-39 (IC), the track sensitivities of samples aged in the freezer were approximately the same as those aged in air at room temperature. From these experimental evidences, we supposed that the ageing effect on CR-39 (IC) was saturated, because these detectors had been supplied to our laboratory in Zagazig after enough sufficient ageing time in air at room temperature before the storage in the freezer. A rapid drop in its initial sensitivity over the first several days for newly manufactured CR-39 has been reported by the Bristol group [9]. Recently, Fujii and his collaborators discovered that the initial rapid drop of the sensitivity of CR-39 was attributed to the escape of CO_2 from the polymer material by diffusion if the detector stored in air at room temperature [16]. Our experiments confirm that the ageing effects on the sensitivity of polymeric track detectors are largely dependent on the storage conditions that affect the diffusion of CO_2 dissolved in the polymer.

TABLE I. The summary of measured data for Intercast CR-39 (IC) and both of homemade CR-39 (HM) and SR-90 (HM) stored in freezer and in room temperature for seven and two years, respectively.

	Detectors aged in freezer			Detectors aged in air at room temperature		
	CR-39		SR-90	CR-39		SR-90
	IC	HM		IC	HM	
Ageing period (months)	84	24	24	84	24	24
Track Height (Å)	197	271	593	204	387	417
Bulk etching rate (μm/h)	1.61	1.61	2.50	1.61	1.61	2.50
Surface Roughness (Å)	29.9	30.0	81.6	38.8	56.3	74.9
Average (Å)	19.9	17.9	41.7	29.5	26.8	41.6
Distance (μm)	0.228	0.294	0.353	0.184	0.137	0.304

4. Conclusions

As can be observed, the ageing of the detection materials can be a source of error on the measuring using the Nuclear Track Methodology. To avoid this problem is recommended to make a pre-etching of the material or a chemical treatment with Methanol solution.

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