TEMPERATURE EFFECTS IN POLYIMIDES IRRADIATED BY HEAVY IONS

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Abstract. The thermal stability of heavy ion latent tracks in polyimides in the temperature range of 100°C to 300°C and the temperature effect during the irradiation in temperature interval of -180°C to 20°C are investigated. The chemical etching of the latent tracks was carried out in H_2SO_4 solutions containing potassium bichromate or in NaOCl (8% Cl) solution. Experimental results showed that tracks in PI are resistant to high temperatures and the track formation is scarcely affected by the temperature during irradiation.

1. INTRODUCTION

Compared with other polymer films, polyimides (PI) are characterized by high thermal and radiation stability and good mechanical, electrical and chemical characteristics. These significant advantages allow one to consider PI as a prospective material for the production of track membranes and nuclear track detectors.

As it is known, the mentioned properties of polyimides are connected with rigid-chain structure of their macromolecules. One can expect that latent tracks in polyimide can also be characterized by high thermal stability and, more generally, by insensitivity to the temperature at different stages of formation and existence of tracks.

In this paper we present the results of two series of experiments performed with the aim to investigate: i) the effects of high temperature annealing on latent heavy ion tracks in PI; ii) the influence of the temperature during irradiation on the etching properties of the tracks.

2. EXPERIMENTAL

The polyimide films - 50 μm thick Kapton (produced by Du Pont) and PM-1 (produced in the former Soviet Union) with the thicknesses of 12 μm and 40 μm - were irradiated by accelerated krypton ions with the energy of ca. 5.5 MeV/nucleon from the GANIL and xenon ions with energy of 1 MeV/nucleon on the U-300 cyclotron. Irradiated films were annealed at different combinations of temperature and time (see Table 1). The exposed films were etched with a solution on the basis of 16N H_2SO_4 and potassium bichromate (50 g/l) at temperatures of 90°C to 100°C. After etching the samples were investigated by a scaning electron microscope JSM-840 (JEOL).

The other series of samples of 7.5 μm thick Kapton films was irradiated by collimated fission fragments from the U-235 target at different temperatures lying in the range of -180°C to 20°C (-180, -170, -160, -150, -140, -120, -110, -70, -50, -30 and 20°C). The uranium target was exposed to the thermal neutron flux (10^8 $cm^{-2}s^{-1}$) from a nuclear reactor. The track density in the samples was ca 10^6 cm^{-2}. The conductivity of the samples was measured at 70°C in the solution of NaOCl (8% Cl) which served as etchant and electrolyte simultaneously. The conductivity data were converted into the

Table 1. Results of the thermal annealing of heavy ion tracks in polyimides

Temperature	Time, h	Type of PI/projectile	Result
170°	18	12 µm PM-1/Kr	yes
		40 µm PM-1/Kr	yes
200°	6	12 µm PM-1/Xe	yes
300°	6	12 µm PM-1/Xe	yes
	6	12 µm PM-1/Kr	yes
	6	50 µm Kapton/Kr	yes
330°	6	12 µm PM-1/Kr	yes/no
	6	50 µm Kapton/Kr	yes/no
360°	3; 6	12 µm PM-1/Kr	yes/no
	3; 6	50 µm Kapton/Kr	pits/no
400°	1.5	12 µm PM-1/Kr	yes/no
	6	12 µm PM-1/Xe	no
	1;1.5;2;3	50 µm Kapton/Kr	yes; pits/no;no;no
	1;2;3;6	40 µm PM-1/Kr	yes/no; no; no; no

values of effective pore diameter. The sample surfaces after etching were observed by SEM.

3. RESULTS AND DICUSSION

3.1. Annealing of heavy ion tracks at high temperatures

It was shown in the paper [1] that Xe tracks in PI seemed to be not faded in the temperature range of 100°C to 300°C at short annealing time (2-3 hrs). In the present work we extended the range of annealing time and temperature. Since we expected complete fading of tracks at the temperatures close to the highest service temperature of PI (400°) the testing of etchability was performed in the 'yes/no' mode. Table 1 illustrates the results of experiments carried out for two types of polyimides, two types of ions and various conditions of annealing. Figures 1 and 2 show the surfaces of PM 1 and Kapton after annealing and etching.

As can be seen from Table 1, partial fading of latent tracks is observed even at 330°C (after 6 hrs annealing of PM-1). Exposure of 50 µm thick Kapton at 360°C during 3 and 6 hours leads to complete disappearance of tracks on one side of the film. On the other side there were observed only surface pits. All types of the examined polyimides showed short conical tracks after the treatment at 400°C during 60-90 minutes. At the longer time of annealing the tracks are faded completely.

The electron photomicrographs (Fig. 1) show that there exist the domains with significantly distinct responses to the annealing. The etched pits having a broad variation of pore diameters and cone lengths are seen on the surfaces of the samples subjected to heating. Along with the tracks strongly faded one can find some tracks which almost were not changed in comparison with the reference sample (photo in Fig.1,a). This may be conditioned by the inhomogeneity of the virgin polyimide [3]. A significant straggling of the experimental data did not allow to measure the parameters of the etching behaviour of annealed polyimides and give quantitative description of the track-fading. But, the overall trend observed for all types of polyimides is the decreasing sensitivity of polyimides with increasing exposure temperature and time (the sensitivity is

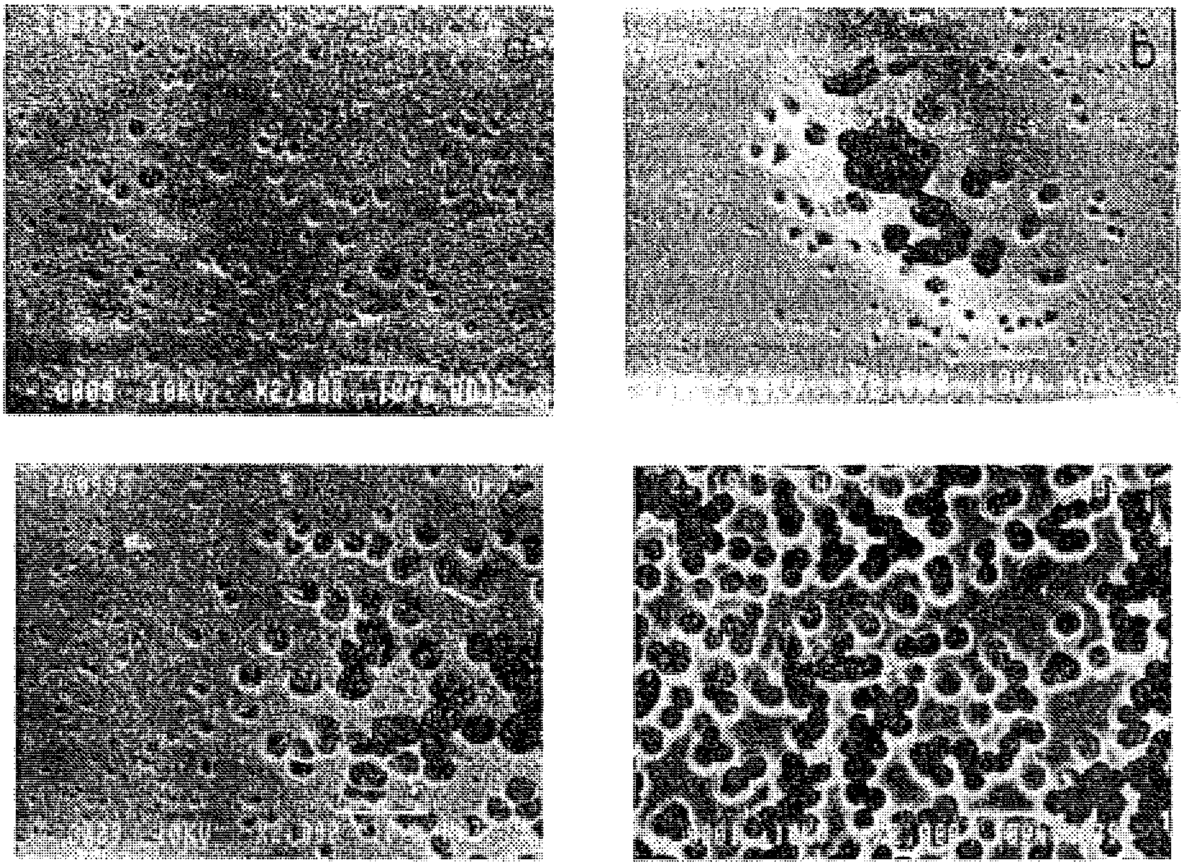

Fig. 1. SEM photographs of the surface of a 12μm thick Pm-1 film annealed at 300° for 6 hrs (a), 360° for hrs (b), 400° for 1.5 hrs (c) and the reference sample (d).

the ratio of track etch rate to bulk etch rate).

3.2. Registration temperature effect in polyimides

During the last decade much work has been done on investigating the effect of temperature during the irradiation with the aim to improve charge resolution of polymeric track detectors [2]. In the present work we investigated a sensitivity dependence on temperature during registration in polyimides. This subject is of interest in respect to the problem of enhancement of the sensitivity of PIs.

The results obtained are shown in Fig.3. The D_{ef} vs t curves have nonmonotonic initial parts which correspond to slow and irregular rise of the conductivity in time. But after about 10-15 minutes of etching the growth of the effective pore diameter is described by a linear function. Linear segments of the D_{ef} vs t dependencies look like regular enlangement of perforated tracks. In order to find out the origin of "irregular" segments on the initial parts of the conductivity curves, the SEM studies of the samples were performed. It was found that under etching the big holes in the polyimide film were developed. The holes are caused by defects having the size close to the thickness of the film used. Some of the defects are being perforated before fission fragment tracks. Only after the etching time long enough the population of particle tracks is transformed to a number of through pores, which grow and contribute to the electrical conductivity.

From the comparison of linear segments of the D_{eff} vs t curves one can conclude that there is no essential difference between the samples irradiated

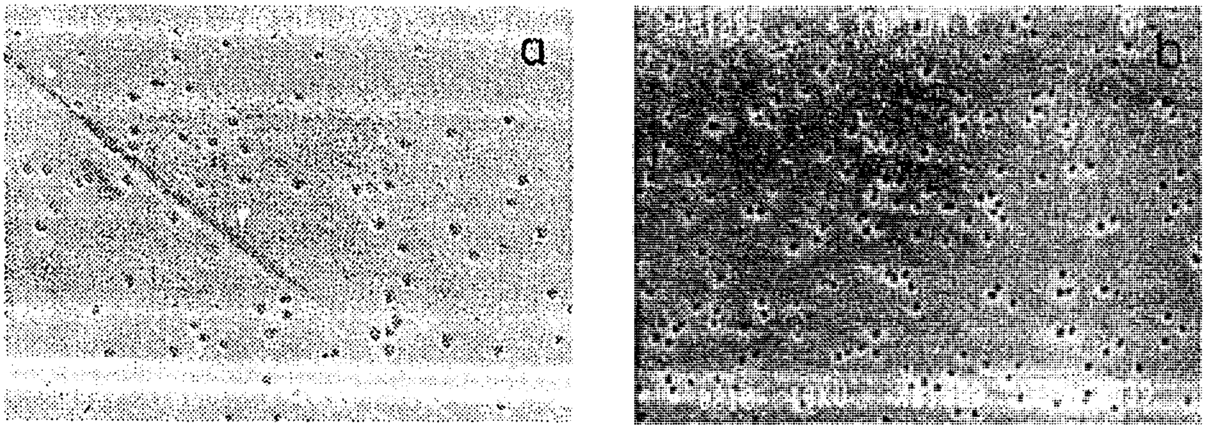

Fig. 2. SEM photographs of the surface of a 50 μm thick Kapton film annealed at 400°C for 1 h (a) and the reference sample (b)

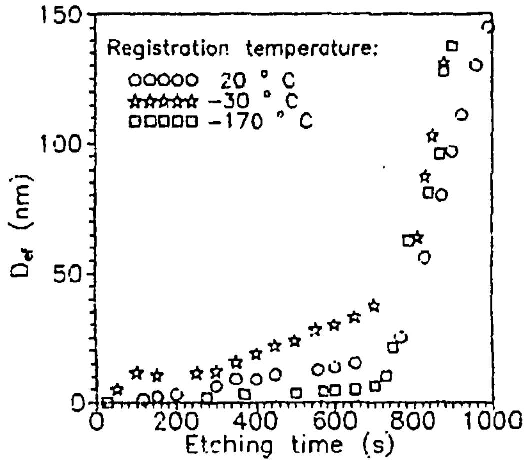

Fig. 3. Effective pore diameter D_{eff} as a function of the time of etching at different registration temperatures

at different temperatures. Therefore, in contrast to the polymers investigated earlier polyimide track detector seems to be insensitive to the registration temperature.

4. CONCLUSION

1. PI seems to be not sensitive to the variation of irradiation temperature in the range of -180°C to 20°C.

2. Heavy ion tracks in polyimide are very stable at elevated temperatures. In comparison with other polymeric detectors polyimides show extreme thermal stability of latent tracks.

3. The response of PI to annealing is essentially inhomogeneous. The manufacture of a polyimide film with homogeneous physico-chemical properties would give, probably, a unique temperature-insensitive track detector.

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