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Optimization of particle fluence in micromachining of CR-39

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Abstract

Polyallyl diglycol carbonate (CR-39 etched track detector) material was irradiated with various doses of 2 MeV protons and alpha-particles in order to optimize the fluence for P-beam writing of CR-39. Irradiation were performed at the Institute of Nuclear Research, Debrecen, Hungary and at the National University of Singapore. Post irradiation work has been carried out in Debrecen. The fluence in the irradiated area was sufficiently high that the latent tracks overlapped and the region could be removed collectively by short etching times of the order of less than 1 min. Theoretical calculations based on analytical and Monte Carlo simulations were done in order to calculate the probability of multiple latent track overlap. Optimal particle fluence was found by minimising the fluence and etching time at which collective removal of latent tracks could be observed. Short etching time is required to obtain high resolution microstructures, while low particle fluence is desirable for economical reasons, and also because high fluences increase the risk of unwanted damage (e.g. melting).

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Keywords: Proton beam micromachining; P-beam writing; CR-39; Polyallyl diglycol carbonate (PADC); Fluence sensitivity

1. Introduction

P-beam writing (or proton beam micromachining, PBM) is an emerging direct-write technique for the production of three dimensional (3D) microstructures in appropriate materials [1–4].

In this work polyallyl diglycol carbonate (PADC) material was selected for target material. PADC (often referred as CR-39) is a thermoset polymer ($C_{12}H_{18}O_7$, $\rho = 1310$ kg/m^3), which is widely used as etched track type particle detector.

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Among the various types of etched track detector materials, PADC has the highest sensitivity for low linear energy transfer (LET) particles, such as protons and energetic cosmic ray particles. LET threshold of this material for individual particles is in the order of 8×10^{-10} J/m at normal particle incidence [5]. Because of its high sensitivity as an etched track detector PADC is also potentially a promising material for PBM as it has already been demonstrated in an our previous work [6]. Its availability in excellent quality from different manufactures is also an advantage for further applications.

Underexposure would result in too rare latent track density, the damaged material would not be collectively removed. Overexposure would increase the risk of unwanted damage (e.g. melting). Thus our aim was to optimize the particle (proton and alpha-particle) fluence in P-beam writing of PADC.

2. Experimental

Tastrak type etched track detector, a PADC (CR-39) material, manufactured by Track Analysis System Limited (Bristol, England) was used in the experiments as a target material. Samples of PADC were cut from large sheets of Tastrak with nominal dimensions of 275 mm × 300 mm × 1 mm. The sample size was 12 mm × 16 mm × 1 mm.

Irradiation of PADC samples with protons and alpha-particles was performed at the nuclear microprobe facility at ATOMKI, Debrecen, Hungary [7–9]. The ion energy was 2 MeV and the beam current varied between 5 and 60 pA. The beam was focused to a spot size of 1–2 μm. The delivered particle fluence was measured using the backscattering signals from the samples by a PIN diode detector array [10].

In an interlaboratory comparison the CIBA ion nanoprobe facility at the National University of Singapore [11] was also used to irradiate samples with a beam of 2 MeV protons focused down to approximately 0.5 μm. The fluence measurements were carried out by measuring the backscattered protons using an annular surface barrier Si detector with an active area of 300 mm^2.

Full square structures with dimensions of 100 μm × 100 μm and 200 μm × 200 μm were irradiated.

After the irradiation, the samples were etched in 6.25 N NaOH solution at 70 °C. Etching was interrupted at every 5 s for monitoring the process of the 3D structure development by optical microscopy observations.

3. Theoretical

In order to achieve collective removal in short etching times, we assume that the samples have to receive such a high fluence of ions, that it causes multiple overlapping latent tracks. The collective removal becomes possible when the overlap is such a high degree that the undamaged fraction of the irradiated region no longer forms an interconnected geometrical network but it forms islands surrounded by damaged material only.

Assume a random areal distribution of latent proton tracks on an area of A. The circular shape cross-sectional area of a single latent proton track is S. At any given point on the area A, the probability that the point is covered k-fold by latent proton tracks is:

$$P_k = \frac{N(N-1)(N-2)\ldots(N-k+1)}{k!}\left(\frac{S}{A}\right)^k \times \left(1-\frac{S}{A}\right)^{N-k},$$

where N is the number of latent proton tracks distributed randomly on the area A. Fig. 1 shows the probabilities of multiple latent track overlap as a function of particle fluence.

A Monte Carlo simulation was also done to calculate the probabilities of multiple latent track overlap. In the simulations the following input parameters have been taken into account: diameter of the single latent track (3 nm for protons, 10 nm for alpha-particles); the beam spotsize (varied between 10 nm and 10 μm); horizontal and vertical beam profiles (Gaussian or trapeziod shape). A typical result is shown on Fig. 2 for 100 × 100 nm^2 beam spotsize and 3 nm diameter proton

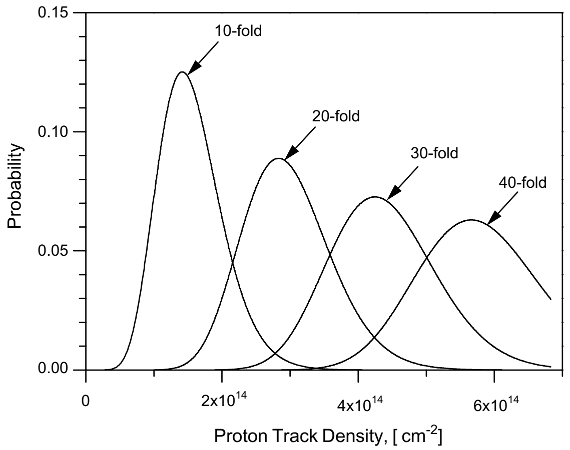

Fig. 1. Probabilities of multiple proton latent track overlap as a function of track density. The diameter of a single latent proton track was assumed to be 3 nm.

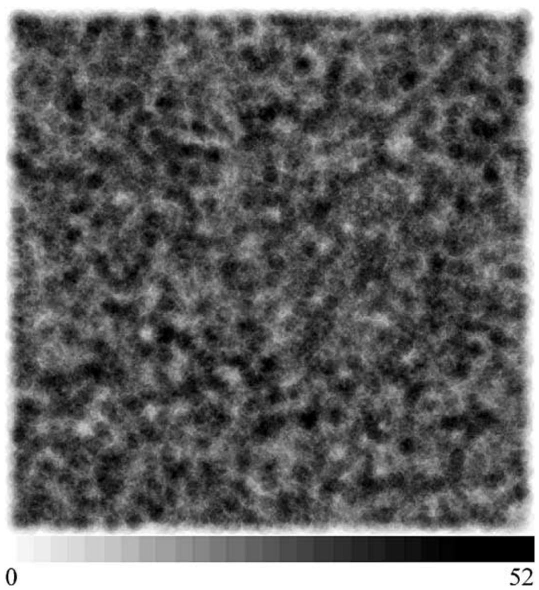

Fig. 2. Grayscale plot (white: minimum intensity, black: maximum intensity) of the multiple latent track overlap based on our Monte Carlo calculations. The square represents $100 \times 100\ \text{nm}^2$ beam spotsize, and a single proton track diameter was 3 nm. The total number of protons was 3.75×10^4 on this surface ($3.75 \times 10^{18}\ \text{m}^{-2}$).

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4. Results and discussion

We have found that the optimum etching time for proton irradiated PADC samples is 60 s. At this etching time the damaged region exposed to the highest fluences were collectively removed, while the ones exposed to the lowest fluences were still not removed at all. Etching times shorter than about 1 min are difficult manage accurately.

The bulk etch rate of the undamaged polymer material is 0.40 nm/s. So the 60 s etching time will result in 24 nm of removed layer thickness, which is also the radius of the roundness of sharp edges. This distance can be considered as a lower limit of spatial resolution of fabricated structures in PADC that can be achieved by PBM. It is nicely below the presently available best beam spotsize [11], while the etching time is still well controllable.

In the applied range of particle fluences we found the optimum fluence for the given etching conditions by analysing the geometrical properties of the etched structures by optical microscopic observations.

Using this method both the Singapore and Debrecen samples gave 600 nC/mm^2 charge density as the best fluence for 2 MeV protons. The irradiated region of the samples was collectively removed and there was a smooth bottom at the end of the latent ion tracks (Fig. 3(b)). At lower fluences the removal was not complete (Fig. 3(a)), and at higher fluences the bottom of the removed pit had an uneven surface (Fig. 3(c)). At the highest applied fluences we observed sample melting. In some cases not only the irradiated but a spherical shape drop-like portion on the bottom side of the irradiated region have been melted, which was removed by etching collectively with the irradiated region (Fig. 3(d)).

The first alpha irradiated samples received 190–2600 nC/mm^2 fluences and they were etched only for 5 s. All irradiated layers were collectively removed, even the one that received the smallest fluence. However, we just managed to see the "lid" (Fig. 4(a)), which means that the etching time for this fluence was on the edge, the lid was also removed after the next 5 s (Fig. 4(b)).

Therefore, we concluded that the CR-39 material is much more sensitive to alpha-particle than

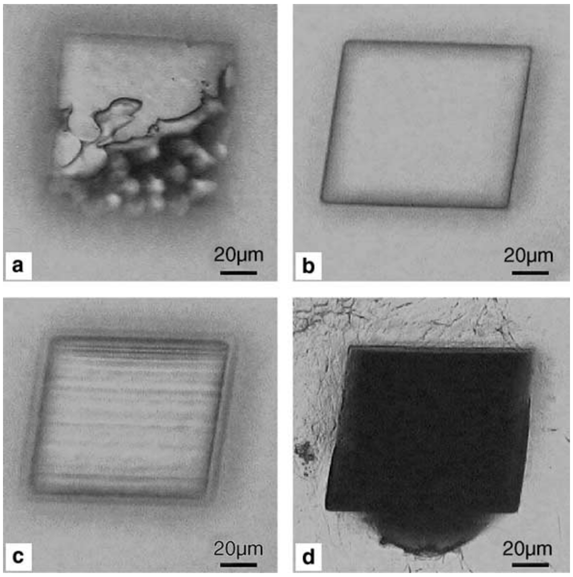

Fig. 3. Proton irradiated samples (a) underexposed sample, partially removed, (b) correct exposure, collectively removed area with smooth bottom, (c) overexposed sample, uneven bottom, (d) overexposed sample, melted area, droplet melted.

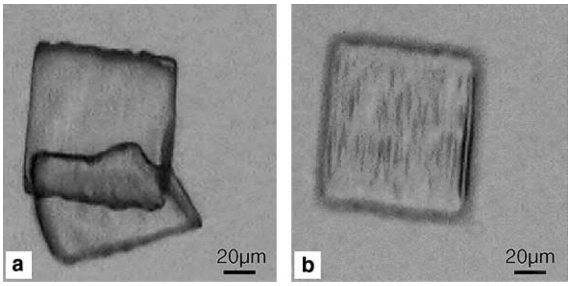

Fig. 4. Alpha irradiated sample (a) after 5 s etching (Note: the "lid" is visible here, the bottom side of it is already out of focus, i.e. it is raised off the surface.) (b) after 10 s etching (the lid is gone, only the removed pit is visible).

for proton irradiation (as it was expected), so we should use lower fluences. Thus a second set of experiment have been performed in which the irradiation dose was 10–300 nC/mm^2. The optimal fluence for alpha particles was found at 60 nC/mm^2.

If one needs very shallow micromachined structures in positive resist, alpha-particle irradiation of CR-39 is a nice solution: it does not need much fluence, and it is more resistant to unwanted damage.

The LET of 2 MeV protons and 2 MeV alpha-particles in PADC is 3.27×10^{-9} J/m and 3.23×10^{-8} J/m respectively [12]. The absorbed dose (D in unit of J/kg) can be calculated by the following formula:

$$D = \frac{\Phi \cdot \mathrm{LET}}{\rho},$$

where Φ is the particle fluence (m^{-2}); and ρ is the density of the material (kg/m^3). Optimal proton fluence was found to be at 3.75×10^{18} m^{-2} (600 nC/mm^2), which results in 9.36×10^6 J/kg absorbed dose at the surface. Similar absorbed dose at the surface was achieved with a fluence of 2 MeV alpha-particles of 3.75×10^{17} m^{-2} (60 nC/mm^2).

Since the ratio of the cross-sectional area of latent alpha and proton tracks is also about 10, this latter value of alpha-particle fluence results in similar multiplicity of latent track overlapping as in case of the optimal proton fluence (according to the above analytical formula and our Monte Carlo simulations).

In comparison, the most suitable range of exposures for 2 MeV protons was found to be 75–85 nC/mm^2 for PMMA [2], and 10–40 nC/mm^2 for SU-8 [13].

5. Conclusions

Polyallyl diglycol carbonate (PADC, also known as CR-39) material has been irradiated with various doses of protons and alpha-particles in order to optimize the fluence for P-beam writing. Optimal fluence for 2 MeV protons was found to be at 3.75×10^{18} m^{-2} (600 nC/mm^2). For alpha-particles the linear energy transfer is 10 times higher. Therefore it is likely that approximately 10 times less fluence is optimal. In our most recent experiment it was found that the optimal fluence for 2 MeV alpha-particles was 3.75×10^{17} m^{-2} (60 nC/mm^2). Low particle fluence is desirable for reasons of efficiency and to avoid unwanted damage, thus alpha-particles outperform protons in P-beam writing into PADC materials.

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