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CREATION OF MICROCAPILLARIES BY P-BEAM WRITING USING DIFFERENT PROTON ENERGIES

G.U.L. Nagy, I. Rajta

Institute for Nuclear Research, Hungarian Academy of Sciences,
H-4026 Debrecen, Bem tér 18/C, Hungary

Abstract

Arrays of circular microcapillaries have been created in Poly(methyl methacrylate) (PMMA) foils using several energies and ion fluences by Proton Beam Writing (PBW) technique. Two series of samples were produced using two different irradiation patterns. The planned total transferred energy was equal for all irradiation energies at a given fluence. The produced capillaries were developed under the same conditions. The effect of the proton energy and the fluence on the capillary size was compared to computer simulations.

I. Introduction

Proton Beam Writing (PBW) is a direct-write microlitographic technique [1-4]. A focused beam of high energy (MeV) ions is scanned over a suitable resist material along a pre-defined pattern creating a latent image. The beam penetrates into the bulk along a straight-line path, because proton-electron collision, which is the most probable interaction between the bombarding ions and the sample, does not cause significant deflection due to the large difference between proton and electron mass. Furthermore, the energy transfer in one collision is small resulting deep penetration of the protons, and the energy deposition along the path is almost homogenous, except for the end-of-range broadening, where nuclear collisions become more prominent (Bragg-peak). This allows the creation of high aspect ratio structures with smooth, vertical sidewalls. A subsequent wet chemical etching process can develop the irradiated pattern and real 3-D structures can be produced.

One of the most common resist materials used for proton beam writing is Poly(methyl methacrylate) (PMMA) [5]. Due to the proton irradiation, the polymer chains are scissioned, making PMMA to be soluble in suitable solutions (developers). The conventional developer used for PMMA is the mixture of isopropanol (IPA) and water in 7:3 ratio. Bolhuis, et al, showed that the GG-developer solution (60% diethylene glycol monobutyl ether, 20% morpholine, 5% ethanolamine, 15% purified water) already used in the LIGA micromachining process is also capable to develop proton irradiated PMMA structures [6].

In this work microcapillary arrays have been created in 50 μm thick PMMA foils with four different energies. Szilasi et al. presented the creation of such capillaries by PBW technique in [7]. These structures may have practical applications in several fields, for example in atomic physics (serving as samples when studying guiding of highly charged ions) or medical and environmental fields (cell filtering, counting, etc.). Huszánk et al. used similar polycapillary films in a microreactor created by PBW technique [8]. The direct write way of PBW provides advantages against conventional filters made by broad beam of heavy ions: the array of the capillaries allows a high ratio of open holes vs. substrate area. In addition, the holes have a circular shape, and they are not overlapped.

The aim of the present work was to compare the effect of the beam energy and ion fluence on the capillary size. We used conventional bright-field optical microscopy for the accurate measurement of the capillary diameters.

II. Experimental

The irradiations were carried out at the MTA Atomki nuclear microprobe facility [9]. 50 μm thick PMMA foils were irradiated with H^+ ions of four different energies: 2 MeV, 2.5 MeV, 3 MeV and 3.5 MeV. The penetration depth of protons with the above energies in PMMA are 65, 93, 126 μm, and 164 μm, respectively, calculated by the SRIM software [10]. Using a 50 μm thick foil, the beam passes the foils at all energies, the end-of-range broadening of the beam (Bragg-peak) is avoided and the protons have nearly homogenous energy loss along their path. The deep penetration allowed us the usage of two 50 μm thick foils placed behind each other in case of the 3 MeV and the 3.5

MeV energy irradiations. Thus the beam energy on the surface of the second sheet was 2.223 MeV and 2.823 MeV, respectively.

In order to reduce irregular fluence distribution over the scanned area due to the beam intensity fluctuations, which is very likely when using belt-driven accelerators, rapid repeated scanning method was used: the total time needed to deliver the desired fluence at a given current was divided by a factor as high as possible but taking care not to result too fast scanning speed, and then the scan was repeated by this number of loops. Thus the intensity fluctuations were averaged over the total area and the capillaries were as uniform as possible over the matrix arrangement.

The irradiation pattern was a 2 x 2 mm^2 square containing an array of ellipses with 11 µm major axes and 7 µm minor axes. It was necessary to plan ellipses instead of circles, because the beam spot was not circular symmetrical, but the vertical spot size was always larger than the horizontal. Two different scanning routes were used for all irradiation energies and fluences (see Fig 1.). The scan resolution was 2048 pixels, thus 1 pixel is around 1 µm. Considering that the vertical extension of the beam is bigger than the horizontal, approximately circular capillaries are expected when major axes are aligned horizontally.

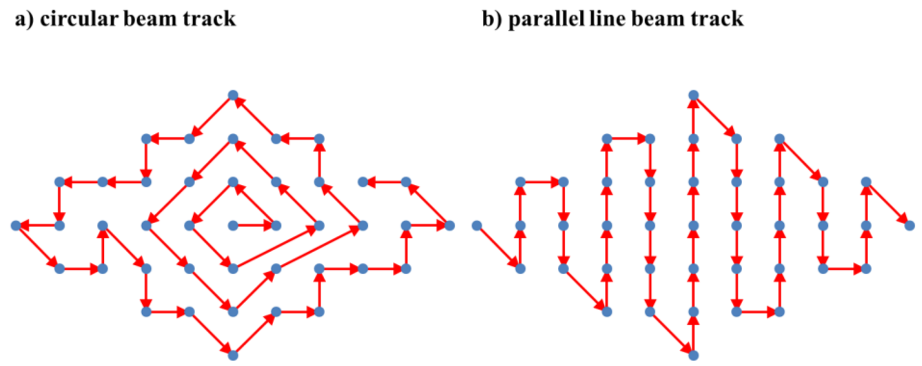

Figure 1: The two different beam routes in one particular capillary. The same pattern was copied near each other for both types until it filled a 2048×2048 pixel matrix.

2 MeV protons transfer 1202 keV total energy during passing through a 50 μm thick PMMA foil. The reference fluence was chosen to be 600 nC/mm^2 and 1000 nC/mm^2 at this energy. Our aim was to achieve the same amount of total adsorbed energy at all irradiation energies. The required fluences for the other irradiation energies to have equivalent total adsorbed energy are summarized in Table 1.

Proton energy (keV)	Transmitted energy (keV)	Lower dose (nC/mm^2)	Higher dose (nC/mm^2)
2000	1202	600	1000
2500	937	770	1283
3000	777	928	1547
3500	677	1065	1775

Table 1: The required fluences at the different energies in order to achieve the same total transmitted energy. The reference dose was chosen to be 600 and 1000 nC/mm^2 for the 2 MeV irradiation.

After the irradiations, the samples were developed in the GG-developer solution (60% diethylene glycol monobutyl ether, 20% morpholine, 5% ethanolamine, 15% purified water). Ultrasonic agitation was used to enhance the developement. At the end of the developing process, the capillaries were ready for the microscopic measurements. The same developing process was used as it was proven good in Szilasi et al. [7] work. I.e. the ultrasonic agitation was performed between two flat glass plates, to avoid sample breaking.

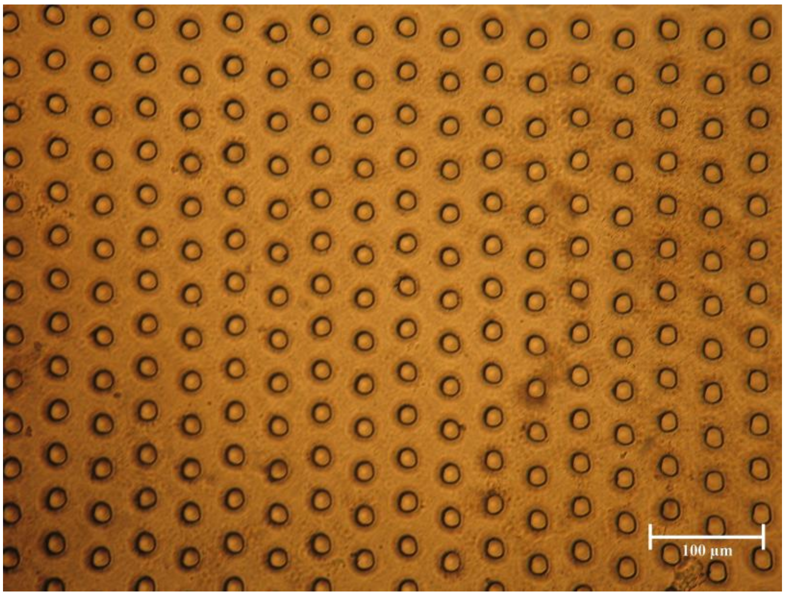

Figure 2: Bright field microscopic image of the capillaries (20× objective; 3 MeV irradiation energy; 1547 nC/mm^2 dose).

III. Results and discussions

The size of the capillaries for all irradiation energies and fluences were measured by conventional bright-field optical microscopy. The results are summarized in Fig. 3.

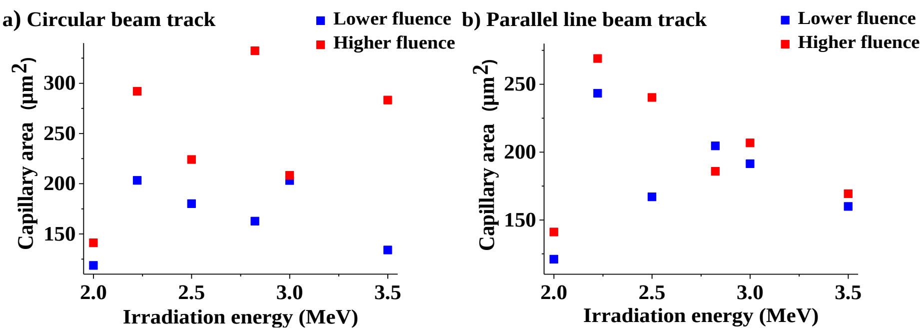

Figure 3: The capillary sizes as a function of the irradiation energy for the two different irradiation patterns.

TRIM (Transport of Ions in Matter, a program included in the SRIM software package) simulations were also performed in order to compare experimental results with theoretical expections (Fig. 4.). The lateral straggling of protons in 50 μm thick PMMA with the energies used in the experiments are summarized in Table 2.

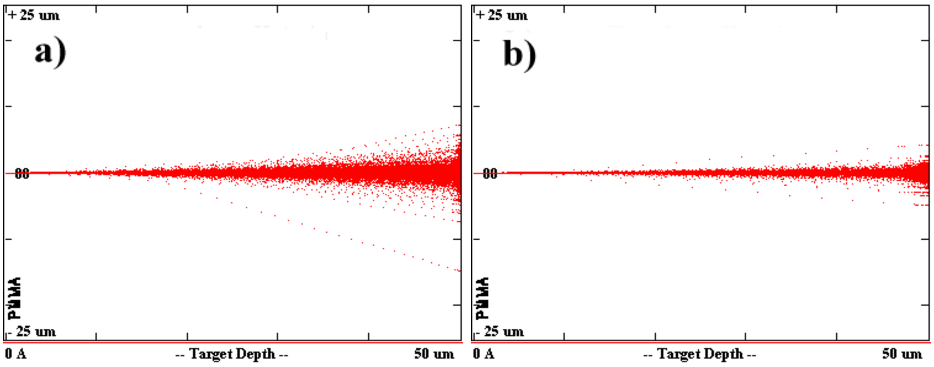

Figure 4: TRIM simulations for the lateral straggling of protons in 50 μm thick PMMA with proton energies of a) 2 MeV and b) 3.5 MeV

Proton energy	2 MeV	2.5 MeV	3 MeV	3.5 MeV
Lateral straggling (μm)	0.67	0.48	0.37	0.31

Table 2: Lateral straggling of protons in 50 μm thick PMMA for the different irradiation energies.

TRIM simulations suggest that with increasing proton energies the lateral straggling of the protons is decreasing. Thus, smaller capillaries are expected for the higher irradiation energies.

The only explanation for the difference between experimental results and theoretical calculations is that the fluctuation of the beam intensity of the old VdG-5 accelerator is as notable that it makes the precise planning of the fluence impossible. In addition, in can be inhomogenous. A higher fluence overexposes the sample, thus the irradiated spot that gave higher fluence than the threshold required to make PMMA developable is bigger than the beam spot size. This is because the beam is assumed to be Gaussian-shaped and the beam size is defined as the FWHM. When the sample is overexposed, the area over the FWHM of the Gaussian beam profile can also get enough fluence to become soluble in the developer.

The new, state-of-the-art 2 MV Tandetron (HVEE) accelerator to be installed at Atomki in the near future will provide proton beams with energies ranging from 200 keV to 4 MeV with orders of magnitudes better intensity stability, according to international experiences. This will help us to the accurate planning of the irradiated fluence.

IV. Summary

Microcapillary arrays have been created in 50 μm thick PMMA foils by Proton Beam Writing technique. Four different proton energies were used: 2 MeV, 2.5 MeV, 3 MeV and 3.5 MeV. The penetration depth of the 3 MeV and 3.5 MeV protons in PMMA allowed us to use two pieces of foils placed behind each other. In these cases the proton energy on the surface of the second foil was equivalent of 2.223 and 2.823 MeV, respectively. For all energies two ion fluences were applied and two different scanning routes were used for both irradiations. The fluences for the 2 MeV irradiations were 600 nC/mm^2 and 1000 nC/mm^2 and the fluences for the other energies were calculated to result the same total transferred energy in the 50 μm PMMA sheet. The irradiated structures were developed in the GG-developer solution, combined with ultrasonic agitation. The size of the capillaries was measured by optical microscopy.

TRIM calculations were also performed to simulate the lateral straggling of the protons in PMMA, and the experimental and theoretical results showed high contrast. The explanation of the results is the bad intensity stability of the old VdG-5 accelerator of Atomki, which makes the accurate planning of the fluence nearly impossible. Much better quality beams are expected from the new 2 MV Tandetron accelerator which will be installed at Atomki in 2014.

Acknowledgements

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