Optically smooth microchannels in the volume of lithium niobate fabricated by selective etching of fs-laser written structures and subsequent annealing

Daniel Nwatu[1], *Detlef Kip*[1], and *Kore Hasse*[1,*]

[1]Faculty of Electrical Engineering, Helmut Schmidt University, 22043 Hamburg, Germany

Abstract. 3D-hollow microstructures with few tens of micrometer in diameter with an optical-quality surface roughness ($R_a \leq 1$ nm) have been fabricated in the volume of lithium niobate by selective etching of fs-laser written structures and post-etching annealing. The fs-laser writing parameters and the annealing process have been refined to reduce the average surface roughness and the shape change. Systematically investigating the annealing process, an empirical functional description of the temporal evolution of the surface roughness was found completing the data set of processing parameters for selective etching of fs-laser written structures, allowing to control the fabrication process of the hollow microstructures concerning both shape and surface roughness. Thus, our results represent another milestone within the research towards monolithic micro-(opto)fluidic applications inside the multifunctional crystal lithium niobate.

1 Introduction

Selective etching of fs-laser written structures is a unique technique to fabricate 3D-hollow microstructures in the volume of transparent optical materials ranging from glasses to crystals such as lithium niobate (LN) [1]. This technique allows to overcome the need for a cover part on top of a structured part containing the microchannels, and thus, paves the way towards monolithic devices [2].

With its piezoelectric, pyroelectric, electrooptic, acousto-optic, and nonlinear properties, which are missing in the common microfluidic platforms like glass or PDMS, LN is a promising multifunctional platform for monolithic micro-optofluidics. Various micro-optofluidic applications have been realized in LN in the past, using a cover piece on a structured bottom containing the microchannels (non-monolithic devices) [3] [4].

In this work, we investigated the post-etching annealing and refined the laser writing and selective etching parameters of LN to achieve an optical-quality roughness of the selectively etched surfaces, what is necessary for micro-optofluidic applications where light (e.g. guided in waveguides) must be transmitted through the walls of the microchannels. We investigated surface smoothening of LN by annealing systematically for the first time, to the best of our knowledge, and found a functional dependence of the average roughness on time and temperature.

2 Experimental methods

For fs-laser inscription a setup described in [1] was used. The laser source, a Femtolux 3, MHz-repetition rate, femtosecond laser with 300 fs pulse duration combined with mechanical stages (XMS-50-S, Newport) allowed for a writing speed of 10 mm/s. Thus, it was possible to structure larger rectangular areas by stacking many lines horizontally with varying distances of 1, 1.5 and 2 µm next to each other (for example, cf. Fig. 1 a)) in two 1×10×10 mm^3 x-cut LN samples.

Before etching in 40% hydrofluoric acid at room temperature, both samples were separated into three parts each so that the acid could reach the laser modified parts of each piece, and to have several similar samples for the investigation of the annealing parameters. After 2 weeks, the channels in all pieces were completely etched. To investigate the surface roughness before and after annealing, three of them were cut open with a precision dicing saw (Disco, DAD322). The other three were used to investigate the change of the cross section during annealing. The samples were annealed at either 1000°C, 1060°C and 1125°C for 5, 25, 50 and 125 h, respectively, measuring the roughness as well as the cross-sectional shape after each annealing step with a laser scanning microscope (LSM, VK-X3000) with a resolution of 1 nm in height.

Additionally, curved and crossing structures were fabricated to show the possibility of fabricating more complex structures in LN by selective etching.

3 Results and discussion

As can be seen in Fig. 1 the structure of the stacked lines during fs-laser writing are still visible on the edges of the fully etched channel (Fig. 1 b)). After only 25 h of annealing the rough edges are completely smooth in Fig. 1 c). The corners get a circular shape during annealing, although the top and bottom surface of the channel remains

---

[*] Kore Hasse: hassek@hsu-hh.de

Fig. 1. Light microscope image of cross section of a) fs-laser modification, b) selectively etched channel, and c) channel after 25 h of annealing at 1060°C.

straight. In case a crystal is heated to a temperature higher than the so-called roughening temperature surface irregularities would start to evolve towards a shape minimizing the surface tension [5]. As a result, the macroscopic roughness would start to decrease. The measured roughness R_a plotted against the annealing time in Fig. 2 can be fitted with an empirical function:

$$R_a = \frac{R_0}{1+a \cdot t^{b\,\exp(-E_A/(K_B T))}}. \quad (1)$$

Here R_0 is the initial roughness before annealing, t is annealing time, E_A is an activation energy, k_B is the Boltzmann constant, T the annealing temperature, and a and b are free parameters. The physical mechanism of the annealing process is not completely understood and although the function above allows to calculate the expected roughness for a certain temperature and time range, detailed computer simulations would be needed to build a solid theoretical model. However, for the targeted application, i.e. fabrication of low roughness hollow microstructures for micro-optofluidic applications, the function above is sufficient. Roughness values down to 1 nm, limited only by the instrument resolution, could be measured. This low roughness, which demonstrates optical surface quality, of microchannel sidewalls in LN could not be reached with other methods [4].

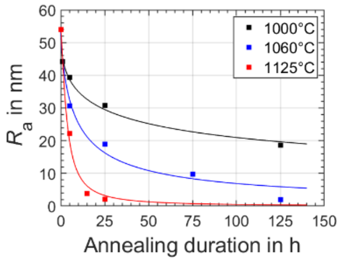

Fig. 2. Measured roughness values plotted vs. annealing duration for an annealing temperature of 1000°C, 1060°C and 1125°C (symbols) and the fitted curves (solid lines) according to Eq. (1). Fit parameters are a = 0.1867, b = 90850, and E_A = 1.337 eV.

As can be seen in Fig. 2, the roughness decreases faster at higher annealing temperatures. Although this would be beneficial for a fast production process, at lower temperatures the shape change during annealing is not so strong compared to higher temperatures. Therefore, for some applications, such as refractive index sensing of liquids using the Fabry-Pérot effect with the channel sidewalls representing the cavity mirrors, lower annealing temperatures would be required, to maintain the highest possible parallelism of the channel sidewalls.

Refining the processing parameters of selective etching of fs-laser written structures in LN, several basic elements of microfluidic channel systems have been realized (cf. Fig. 3), illustrating the method's application potential. The 180° turn in part a) with a radius of 0.62 mm has a regular shape, i.e. the orientation dependence of the selective etching process does not show a significant influence. In Fig. 3 b) such influence is indeed visible. The channel etched along the y-axis is slightly broader than along z due to the higher etch rate along the z-direction of the unmodified material. This effect could be compensated by a broader material modification along the z-direction. In Fig. 3 d) the channels were etched from several stacked fs-laser written lines to achieve a broader channel. Within this channel, droplets, which would form when liquid is brought in contact with the channel entrance, can be recognized even without annealing. Obviously, the liquid was sucked into the channel by capillary forces.

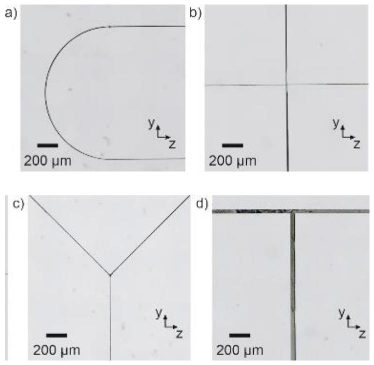

Fig. 3. Microscope images of selectively etched structures in LN: a) 180° turn, b) cross, c) y-junction, d) t-junction etched from stacked fs-laser written tracks to obtain a broader channel cross section. For the latter example, water droplets sucked into the channel by capillary forces can be seen in the microchannel.

4 Conclusion and outlook

A functional relation between surface roughness and annealing temperature and time has been found for the post-etching annealing process. By annealing, the microchannels' sidewall roughness can be reduced down to 1 nm. Additionally, various hollow microstructures were fabricated, such as 180° turnes, y- and t-junction, etc. These results represent a milestone on the way towards monolithic micro-optofluidic applications in LN. More details on both methods and results, including a first demonstration of an integrated Fabry-Perot cavity for refractive index measurements, will be presented at the conference site.

References

1. D. Nwatu, D. Kip, and K. Hasse, Opt. Express **31**(23), 37618–37629 (2023).
2. R. Osellame, G. Cerullo, and R. Ramponi, *Femtosecond Laser Micromachining Photonic and Microfluidic Devices in Transparent Materials* (Springer, 2012).
3. G. Bragato et al., Adv. Mater. Interfaces **2024**, 2301008 (2024).
4. G. Bettella et al., Micromachines **8**(6), 1–13 (2017).
5. W. Selke and J. Oitmaa, Surf. Sci. **198**, L346–L352 (1988).