Pyrolyzed 3D compound refractive lens

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Abstract. X-ray synchrotron and laboratory source radiation is used to solve a wide range of problems in modern science. Compound refractive lenses (CRLs) technology is a popular solution for focusing x-ray. However weak interaction of X-ray radiation with matter puts extra challenges for microfabrication techniques and materials. Two-photon lithography has been successfully applied for polymer CRL microfabrication. However polymer material is likely to degrade in intense X-ray beams. This article describes pyrolysis (heating in inert atmosphere) as an improvement of two-photon lithography technique. This extra step may produce ultrasmall curvature radii CRLs with extra durability in intense X-ray beams

1. Introduction

The combination of X-ray analytical techniques with X-ray coherent micro- and nanobeams and brilliant synchrotron radiation sources may become a unique characterization tool for broad range of scientific tasks in the fields of solid-state and, high-pressure physics, chemistry, biology and cultural heritage. The focusing of X-rays may be achieved by either their reflection (Kirkpatrick-Baez mirror), diffraction (Fresnel zone plates, multilayer Laue lenses) or refraction (CRLs) [1]. CRLs focusing optics finds its applications due its compact size, wide range of operation (10-100 keV) and wide range of accessible focal distances. CRL operate in direct parallels to the visible range refractive optics. However, in X-ray range the refractive index decrement is very small \(1-n \sim 10^{-5}\). Thus, single lens is unable to bend X-ray wavefront sufficiently. Several individual lenses put in an array are required to produce X-ray micro- and nanobeams and form a CRL. The two main types of CRL include Be and Si lenses.

Be CRLs combine a large physical aperture with high transmission. As a result they are able to deliver high flux to the sample. The best reported resolution of Be CRLs achieves approximately 100 nm [2,3]. However these lenses are not very popular to produce nanobeams due to their limited resolution and the presence of aberrations [3].

Higher spatial resolutions have been achieved with Si planar lenses, so-called nanofocusing lenses (NFLs), which are manufactured by planar structuring of silicon. Beam sizes of 50 nm and below are achieved routinely [4-6]. Despite the high resolution Si NFLs have a drawback: their relatively high absorption in Si is limiting the total flux in the nanobeam. Total flux is even more reduced due to technologically limited physical aperture and the use of crossed geometry.

Two-photon absorption lithography is another method used for CRLs fabrication. It combines full geometrical freedom with 100-nm resolution and has been successfully applied for microfabrication of CRL from polymer material [7-9]. However, polymers are likely to degrade in intense X-ray beams leading to deterioration of focusing performance [10]. Slow heating in inert atmosphere (pyrolysis) leads to elimination of the most organics in polymer material. Either carbon or ceramic-type material is
formed [11,12]. Polymer-derived materials show excellent radiation and thermal stability and oxidative resistance [13-15]. Here we present the combination of two-photon lithography technique with subsequent pyrolysis step to produce ultrasmall radii stable CRL.

2. Experimental section

The structure was produced by standard two-photon absorption lithography technique. The ORMOCOMP photoresist from Micro Resist Technology GmbH was used for microfabrication. The molecular formula of ORMOCOMP is C21SO8SiH36. The radiation from Ti:Sapphire femtosecond laser (800 nm, 50 fs) was focused by immersion oil objective (NA=1.4). The exposure parameters were 30 mW of average incident power and 1200 μm/sec beam waist speed. The exposure of the resist was performed from top to bottom in a cell containing uncrosslinked photoresist. The thickness of the cell was approximately 120 μm and was controlled by two pieces of adhesive tape. The printing process was conducted in a layer-by-layer manner with linear infill and 3 perimeters. The slicing and hatching distances were 200 nm.

The aperture of individual lens before pyrolysis was 28 μm and the curvature radius of parabolic profile was 5 μm. The lens was put on 4 legs to ensure isotropic deformation and uniform reduction of lens size (Fig 1 a,b).

For pyrolysis a tube furnace was used. According to [16] the pyrolysis process consisted of 4 steps in order to both evaporate the remaining solvents, as well as to initiate the process of organic decomposition without prominent distortions and as well as to eliminate all organic residues from the
The structure was baked at 690 °C under a constant argon flow. In the first step the temperature was set to 250 °C for 60 minutes. Then the temperature was raised to 350 °C at 5 °C/min and the structure kept at this temperature for 60 minutes. Finally the temperature was set to 690 °C at 5 °C/min and the structure was kept at this temperature for 1 hour. Finally, heating was switched off and the structure was left to cool down under constant argon flow.

The microstructure was characterised by scanning electron microscopy before and after pyrolysis (Fig. 1 c,d). The process of pyrolysis lead to size reduction in approximately 2.3 times. We used energy dispersive X-ray spectroscopy (EDX) to monitor changes in chemical composition of the material. The EDX system were coupled with scanning electron microscope. The normalised intensities of EDX signals are presented in table 1. Heating in inert atmosphere of the microstructure leads to drastic changes in chemical composition: the pyrolyzed microstructure consists mainly from Si and O.

|                          | C, arb units | Si, arb units | O, arb units | S, arb units |
|--------------------------|-------------|--------------|-------------|-------------|
| before pyrolysis         | 58          | 4            | 35          | 3           |
| after pyrolysis          | 3           | 27           | 70          | 0           |

*Table 1 Normalised intensities of energy dispersive X-ray spectroscopy signals for C, Si, O and S.*

The change in chemical structure, density and geometry of individual lens will lead to the change of X-ray optical properties and as a result focusing performance of a single lens: we expect the refractive index decrement to shift from 2.69 10⁻⁴ to 5.54 10⁻⁴ and linear attenuation coefficient from 0.97 10⁻⁴ µm⁻¹ to 4.9 10⁻³ µm⁻¹ at 10 KeV. We expect the focal distance of single lens to shift from 93 cm to 20 cm at 10 KeV.

3. Conclusion

A new approach for 3D compound refractive lenses production was presented. The pyrolyzed lenses consist mainly from SiO2 which presumably would make them more resistant to x-rays. Concomitant size reduction of 2.3 times makes this approach perspective for extra small radii of curvature lenses fabrication. Further research to validate these claims will be required.

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4. References

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