LETTER

Femtosecond laser-induced stress-free ultra-densification inside porous glass

To cite this article: Vadim P Veiko et al 2016 Laser Phys. Lett. 13 055901

View the article online for updates and enhancements.

Related content

- Enhancing nonlinear energy deposition into transparent solids with an elliptically polarized and mid-IR heating laser pulse under two-color femtosecond impact
  F V Potemkin, E I Mareev, Yu I Bezushnova et al.

- The pulse duration dependence of femtosecond laser-induced refractive index modulation infused silica
  Hengzhang Guo, Hongbing Jiang, Ying Fang et al.

- Selective cell adhesion on femtosecond laser-microstructured polydimethylsiloxane
  A M Alshehri, S Hadjiantoniou, R J Hickey et al.

Recent citations

- Ultrafast Laser Pulses for Structuring Materials at Micro/Nano Scale: From Waveguides to Superhydrophobic Surfaces
  Daniel Correa et al

  Jarlath McKenna

- Enhancing nonlinear energy deposition into transparent solids with an elliptically polarized and mid-IR heating laser pulse under two-color femtosecond impact
  F V Potemkin et al
Femtosecond laser-induced stress-free ultra-densification inside porous glass

Vadim P Veiko\textsuperscript{1}, Sergey I Kudryashov\textsuperscript{1,2}, Maksim M Sergeev\textsuperscript{1}, Roman A Zakoldaev\textsuperscript{1}, Pavel A Danilov\textsuperscript{2}, Andrey A Ionin\textsuperscript{2}, Tatiana V Antropova\textsuperscript{3} and Irina N Anfimova\textsuperscript{3}

\textsuperscript{1} ITMO University, Kronverkskiy prospect 49, 197101 St Petersburg, Russia
\textsuperscript{2} Lebedev Physical Institute, Leninskiy Prospect 53, 119991 Moscow, Russia
\textsuperscript{3} Grebenshchikov Institute of Silicate Chemistry, Russian Academy of Sciences, 2 Makarova nab., 199034 St. Petersburg, Russia

E-mail: zakoldaev@gmail.com and sikudr@sci.lebedev.ru

Received 22 January 2016, revised 3 March 2016
Accepted for publication 9 March 2016
Published 1 April 2016

Abstract
Unusually high densification \(<26\%\) was obtained without lateral residual stresses within the laser beam waist inside porous glass during its multi-shot femtosecond laser irradiation, which may induce in the glass the related high refractive index change \(<0.1\). Corresponding laser irradiation regimes, resulting in such ultra-densification, decompaction and voids inside the glass, were revealed as a function of laser pulse energy and scanning rate, and were discussed in terms of thermal and hydrodynamic processes in the silica network.

Keywords: nanoporous glass, femtosecond laser pulses, densification

(\textit{Some figures may appear in colour only in the online journal})

1. Introduction

Porous glasses (PG) represent a specific promising kind of glass with multiple buried hollow channels and pores, which can be potentially filled in with different substances to provide selective absorption, lasing, and optical nonlinearity [1–3]. Typically, an average pore size in PG samples varies from 2 to 10 nm [4]. Local variation of PG mass density allows the fabrication of different complex embedded 3D-microstructures—aspherical micro-optical elements, matching components or soft diaphragms [5–9]. Laser-induced variation of local PG density up to complete closure of pores makes it possible to vary broadly adsorption properties of PG, thus providing control of diffusion processes in such a branched system [10–13]. On the other hand, local decompaction up to the formation of hollow micro-channels in some pre-defined configurations with pre-determined channel diameters is also highly demanding. As a result of such local densification and decompaction laser-micromachining processes, PG is a novel emerging optical platform for the fabrication of integrated micro- and nano-devices for photonic, plasmonic, microfluidic and micro-pneumatic applications [14–18].

Similar changes of local mass density and the related refractive index in different bulk silica glasses (in particular, fused silica), induced by ultra-short laser pulses, have attracted a lot of attention in previous years [19–22]. On this material platform, different buried optical and microfluidic elements—waveguides, chambers, channels, their circuits or integrated microdevices can be routinely fabricated via laser-induced 3D local micromodification of mass density and related refractive index in such materials. Unfortunately, only minor refractive index changes were be achieved in this way (\(\Delta n \sim 10^{-3}–10^{-2}\)), owing to intrinsic mechanical and energetic limitations on densification and decompaction in such normal-density materials [23]. This strongly contrasts with porous glass as a novel emerging optical platform for fs-laser micromachining, where much larger local changes in mass density and the related refractive index (up to \(\Delta n \sim 0.12\) in the quartzoid phase) become possible [24]. Meanwhile, up until now, the only local surface PG densification with the refractive index change from 1.33 till 1.46 has been accomplished under the action of the CO\textsubscript{2} (\(\lambda = 10.6\ \mu\text{m}\)) laser radiation [6, 8, 25] over the 25–50 \(\mu\text{m}\)-wide modification regions, owing to the commonly strong CO\textsubscript{2}-laser absorption in silica glasses [26].
Moreover, local density changes inside PG may potentially proceed without the appearance of lateral residual stresses [9], which are usually present for normal-density glasses [27]. Hence, the highly promising PG potential for fs-laser microfabrication of diverse advanced micro-photonic devices has not yet been explored.

In this Letter, we report on fs-laser irradiation regimes, resulting in stress-free ultra-densification, decompaction and void formation inside porous glass as a function of laser pulse energy and scanning rate, and discuss these regimes in terms of thermal and hydrodynamic processes.

2. Experimental details

We used PG samples (8V-NT type) in the form of 1 mm thick polished plane-parallel plates. The average pore size was 5 nm, the total porosity $\sigma = 26\%$ and the specific surface of the pores $\approx 210 \text{ m}^2\text{g}^{-1}$. The chemical composition was (mass fraction, %): $0.30 \text{Na}_2\text{O} - 3.14 \text{B}_2\text{O}_3 - 0.11 \text{Al}_2\text{O}_3 - 96.45 \text{SiO}_2$ and the expected trace content of $\text{Al}_2\text{O}_3 \leq 0.1$ mass % [28]. The plates exhibited high transmission $\sim 90\%$ in the visible and near-IR range ($0.4-2.5 \mu m$) with the effective refractive index $n \approx 1.34$.

Laser micromachining inside the PG samples was carried out, utilizing a Yb-doped fiber laser Satsuma (Amplitude Systèmes, France) at the 515 nm second-harmonic wavelength, the pulse duration (FWHM) of 200 fs, the maximum pulse energy $E_p \approx 2.3 \mu J$ and the fixed repetition rate of 500 kHz. The samples were arranged onto a computer-controlled XYZ translation stage Standa (8SMC1-USBhF), substituting a sample stage of a microscope Levenhuk 630, and linearly translated at different scan velocities $v_1$ in the range of $0.01-4 \text{ mm s}^{-1}$. A 10 × objective (NA = 0.25) of the microscope focused the fs-laser pulses introduced via its upper trinocular input, inside the PG samples, while an ocular color videocamera DCM310 was arranged in a binocular set to monitor the micromachining process. The schematic view of the experimental arrangement is shown in figure 1. At the focusing conditions, the pulse energies $E_p = 1.5-2.3 \mu J$ of femtosecond laser pulses were sufficient to achieve in the beam waist corresponding peak laser intensities of $8.0 \cdot 10^{13}-1.2 \cdot 10^{14} \text{ W cm}^{-2}$.

3. Experimental results and discussion

Different fs-laser micromachining regimes were observed in our experiments, when the basic experimental parameters — scan speed and laser pulse energy — were varied in the ranges, represented in table 1.

Different linear microstructures were produced inside the PG samples, scanned at variable speeds, as a result of femtosecond laser pulses micromachining at different pulse energies. In the first regime, bright linear light-focusing ultra-densification regions with the related increased refractive
index appeared for \( E_p > 1.5 \, \mu J \) in the speed range \( v_1 = 0.625-3.75 \, \text{mm s}^{-1} \) (figure 2(a)). Images of the buried regions inside the PG sample were also taken, using linearly polarized light with a crossed polarizer/analyzer pair (figure 2(b)), indicating the absence of lateral residual mechanical stresses around the densified region.

In the second regime, the increased number pulses per shot at the lower scan speeds \( v_1 = 0.05-0.625 \, \text{mm s}^{-1} \) and the slightly higher pulse energy \( E_p \approx 2.2 \, \mu J \) resulted in local partial decompaction inside the PG sample (the dark defocusing or scattering regions in the transmitted light in figures 2(c) and (d)). Compared to the first regime, the size of the laser-modified regions in this regime increased by two times in response to just a 10%-increase of the fs-laser pulse energy, indicating the threshold-like character of decompaction in terms of \( E_p \). Nevertheless, images of the laser-decompacted regions in linearly polarized light with a crossed polarizer and an analyzer demonstrated no collateral residual stresses.

Finally, laser micromachining in the third regime at the higher pulse energy \( E_p \approx 2.3 \, \mu J \) and the lower scan speed \( v_1 = 0.012 \, \text{mm s}^{-1} \) yielded in the formation of voids, overlapping during the multi-shot PG exposure in the form of hollow channels (figure 2(e)). Following the even smaller (5%) increase in \( E_p \), the lateral size of the laser-modified regions increased by five times—specifically, from 3 to 15 \( \mu m \), while, again, lateral residual stresses were absent (figure 2(f)).

Overall, these three experimental regimes are presented in figure 3 in the coordinates ‘laser pulse energy—the number of accumulated laser pulses’. This figure demonstrates their well-defined regions with the prominent borders in the parameter space, where both fs-laser pulse energy and the number of accumulated pulses are crucial to manage thermal, hydrodynamic and mechanical dynamics in the molten material.

In terms of fs-laser pulse energy, the appearance of distinct ultra-densified regions in the form of waveguide fragments (figure 2(a)) for \( E_p > 1.5 \, \mu J \) can be related, according to previous calculations [29, 30], to PG heating in the focal region via nonlinear optical effects [24] till its densification temperature \( \approx 1 \times 10^3 \, ^\circ\text{C} \), when pores collapse inside the glass [31]. Below the densification threshold energy, only occasional separate densified regions can be observed during scanning of the PG bulk by the focused fs-laser beam owing to its intrinsic inhomogeneities. Densification of this material in the focal region makes possible viscous hydrodynamic flows, gradually—from pulse to pulse—rising to rather homogeneous densification in the focus without lateral stresses (figure 2(b)), which relax over the molten or softened peripheral material via nanoscale growth of the intrinsic pores. It is the nanoporous PG character that provides its moderate densification without residual lateral stresses. In addition, the size of densified regions gradually changes from 3 to 5 \( \mu m \) versus increasing the number of pulses accumulated inside the micromachining area (figures 3(a) and (b)).

Then, at higher fs-laser pulse energies—up to 2.2 \( \mu J \)—the temperature of the molten glass in the focal region can approach \( 3 \times 10^3 \, ^\circ\text{C} \) [29, 30], exceeding the silica boiling temperature \( \approx 2.2 \times 10^3 \, ^\circ\text{C} \) [32]. Such higher temperatures of the molten glass are favorable for its faster—for the smaller number of pulses—ultimate densification to the quartzoid density of 2.2 g cm\(^{-3}\) owing to the reduced melt viscosity [33]. In contrast, the pronounced reduction of the refractive index is observed for \( E_p > 2.2 \, \mu J \) (dark track of light-defocusing voids in figure 3(c)), indicating the central rupture of the densified core along the scanning direction (local decompaction, void formation). Such decompacted microwebspaces were recently studied in detail [19, 34, 35], revealing their nanoscale heterogeneous (foam-like) structure and possible formation mechanism [36]. Possible reasons for such rupture were considered to be non-threshold residual lateral stresses in quenched silica melt [34] or a threshold-like onset of homogeneous glass boiling in the
central temperature maximum and the related thermoacoustic generation of radial compression and succeeding rarefaction pressure waves, with the latter one potentially initiating boiling and rupture in the center [34]. The nanofoam-like morphology of the decompacted region in figure 2 in [34] supports the tension/boiling decompaction origin, similarly to the spallative rupture on the fs-laser ablated surfaces [37].

Finally, at the maximum pulse energy $E_p = 2.2 \mu J$ and the high number of accumulated laser pulses $N \sim 10^6$, separate decompacted regions as chain-like sequences of voids (figure 3(d)) merge into a continuous hollow channel (figure 3(e)). At the lower scanning speeds, the large number of pulses accumulated per spot acts as effective upscaling of melt lifetime during decompaction, supporting void formation and merging during slow viscous hydrodynamic flows. As a result, the channel diameter gradually rises till 15 $\mu m$ for the increasing number of accumulated pulses (figure 3(e)).

4. Conclusions

In conclusion, we studied femtosecond laser micromodification processes in porous glass, which is a novel promising optical material platform for fabrication of integrated photonic micro- and nano-devices. A gradual sequence of ultra-densification, decompaction and void formation processes was observed in such glass via its single scan by varying energy and/or the number of accumulated focused femtosecond laser pulses. It is the porous mesostructure of the material that enables its ultimate densification and, potentially, the related high refractive index contrast. Moreover, residual lateral tensile or compressive stresses can be nearly completely eliminated (damped) by the porous structure upon ultra-densification or hollow channel formation, respectively.

Acknowledgment

This work was supported by the Russian Science Foundation (agreement № 14-12-00351) and by the Program of Basic Scientific Researches of the State Academies of Sciences (agreement N 0097-2015-0021).

References


