fs-laser induced transformations in oxide glasses for high temperature photonic applications

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Abstract

Femtosecond (fs) lasers enable high peak powers and local transformations thus allowing 3D processing of transparent materials, such as silica and silicate optical fibers or bulk glasses. Therefore, fs-laser direct writing (FLDW) is an attractive tool for a variety of applications in glass science including, among others, micro-optical phase elements and birefringent devices, microfluidics, or long-term optical data storage. Over the last years a strong interest has grown for high temperature sensing for structural health monitoring, steel and metal industry, nuclear power plants or engine gas turbine combustion for aircrafts.
In this context of high temperature sensing (typ. $>$ 800 ◦C for years), one must imprint photo-induced transformations in glass that can be stable, i.e., do not erase or evolve over time, at the device time and temperature operating conditions. In silica, there exist three main laser transformation regimes, commonly labeled Types I, II, and III. Type I is characteristic of an isotropic refractive index variation upon irradiation. Part of this refractive index increase is related to defect formation and densification, which tends to erase within few hours at / or below 500 ◦C. Therefore, type I is not suitable for high temperature environments. Types II and III, on the other hand, allow much higher thermal stability (typ. 1000◦C or slightly higher temperatures for 100's of hours). Type II regime corresponds to the formation of a refractive index sub-wavelength modulation, while type III relates to the formation of void-like structures. Although these two types of transformations are of interest, Type II brings additional degree of flexibility with respect to type III. The so-called nanogratings, typically formed in the type II regime, are birefringent nanostructures, constituted of sub-wavelength porous nanoplanes. They are sensitive to light polarization, which is an asset for many photonic applications.

However, to date, the influence of glass composition on the thermal stability of such nanogratings is not fully understood and would ultimately cause drifts in the envisioned sensor applications when monitoring a given optical response, for example in Fiber Bragg gratings or Fabry-Pérot devices. Recently, using both bulk glasses and optical fibers, we identified that glass viscosity, and its evolution as a function of temperature, are key players in the extraordinary thermal stability of the Type II modifications. Moreover, viscosity also plays an important role on the ability to induce nanogratings in multicomponent glasses. These two aspects will be first discussed at the conference. Both the impact of laser parameters (e.g., pulse energy, writing speed) and glass composition (network modifier, B2O3 and Al2O3 concentration) will be considered. Moreover, the experimental evolution of a polarization

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sciencesconf.org:ustv-dgg-2023:447123

dependent birefringence, which is a characteristic optical signature revealing the presence of nanogratings, will be compared to a theoretical model (Rayleigh-Plesset) to support the findings.

While the aforementioned approach is satisfactory, we recently observed that some glass compositions (Al2O3-SiO2 glass with Al2O3 $\approx$ 50 mol%) deviated from the above trend, i.e., the glass thermal stability is no longer function of the glass temperature-dependent viscosity. These glass compositions can be fabricated using nonconventional techniques, i) aerodynamic levitation for bulk samples and ii) molten core method for optical fibers. Interestingly, under similar annealing conditions, the nanogratings erasure temperature is found to be higher in these compositions with respect to either pure silica (Suprasil F300, or Ge-doped SiO2, the latter being typically found in conventional fiber core compositions). While these demonstrations may offer an exciting path towards enhanced thermal stability performance in glass fiber-based devices, one must comprehend the underlying mechanisms dictating this unexpectedly high thermal stability. From this remark, we recently investigated a 60%Al2O3-40%SiO2 bulk glass sample composition and probed the laser parameter versus thermal stability landscape. Laser pulse energy, pulse duration, writing speed, laser polarization, repetition rates were varied, and isothermal annealing experiments performed, during which monitoring of optical retardance were carried out.
Following the trend deviation from the 50%Al2O3-containing glass, we observed that retardance could still be detected after 30 min annealing at 1400 $\circ$C in these a 60%-Al2O3 containing glasses, well above SuprasilCG (1200$\circ$C) serving as a reference. Moreover, this unexpected stability was observed for most of the probed laser parameters. Micro-Raman analysis on laser-track cross sections showed that both molecular oxygen, characteristic of porous nanogratings, along with mullite crystallization, were detected in the laser-modified area. These results will be presented at the conference and support the claim that photo-precipitation of refractory nanocrystals can be a viable route to induce high-temperature resistant modifications inside a glass.

Keywords: Photosensitivity, femtosecond laser, silicate glasses, high temperatures