
Relaxation processes of densified silicate glasses having different thermo-mechanical histories

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Abstract

Depending on the thermo-mechanical history, SiO₂ glass samples, with same density, exhibit structural differences (ring population, intertetrahedral angles, ...). From these results, it's interesting to wonder about the role of pressure and temperature during the densification process. One possibility, little studied in the literature, to more understand these structural and thermodynamic differences, is to study the relaxation mechanisms during a temperature annealing.

We have studied relaxation during different high temperature annealing, far below glass transition temperature, densified silicate glasses in order to more understand structural processes occurring during annealing for both pure SiO₂ and Na₂O-SiO₂ glasses. For that, we have prepared several recovered densified samples from Belt press (High temperature and high pressure) and from diamond anvil cell (room temperature, high pressure). In-situ experiments (Raman spectroscopy, X-ray diffraction, small angle X-ray scattering (SAXS)) have been done during annealing.

Our results put in light that, during temperature annealing, the density decreases monotonously while the structural evolution shows a non-monotonous behaviour. Indeed, both density fluctuations from SAXS and D₂ band area, related to 3-fold ring population, from Raman spectroscopy first increase before decrease during the temperature annealing. Then, the relaxation occurs from a transitory state which leads to an inhomogeneity increase during the process. Moreover, the role of sodium cations leads to a more compact glass with a decrease of the free volume and an increase of disorder. Nevertheless, in the sodo-silicate glasses, the transitory state has been also put in evidence during the relaxation process of these recovered densified glasses.

Keywords: Silicate glasses, vibrational spectroscopies, high pressure, temperature relaxation, small angle X, ray scattering, structure

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