Densification and relaxation of densified glasses under electron irradiation in silica and phosphate glasses

Nadège Ollier*¹, Matthieu Lancry², Mohamed Mahfoudhi , Sebastien Maron , and Daniel Neuville³

¹Laboratoire des Solides Irradiés (LSI) – Ecole Polytechnique, Université Paris Saclay – France
²Institut de Chimie Moléculaire et des Matériaux d'Orsay (ICMMO) – Université Paris Saclay – France
³Institut de Physique du Globe de Paris - Sorbonne Paris Cité (IPGP) – IPG PARIS, CNRS :
UMR7154 – 1, rue Jussieu - 75238 Paris cedex 05, France

Abstract

It is possible to permanently densify oxide glasses under High Pressure (HP) either at room or high temperature. In silica glass, electron irradiation can also lead to a density increase but it is less efficient (a few % against 20% under HP). In this work, we are interested in the density variations of phosphate and silica glasses under 2.5 MeV electron irradiation. A significant density increase (up to 13%) was obtained in Zn metaphosphate after a 4x109 Gy integrated dose. Note this value is comparable to what was obtained under 2 GPa at Tg [1]. The increase of glass density will be analyzed in different metaphosphate and polyphosphate glasses as a function of the integrated dose, the initial polymerization degree of polyphosphate glasses as well as the presence of Zn. This will be discussed in terms of structural variations analyzed by Raman spectroscopy and solid state NMR of 31P. In pre-densified silica glasses either by HP-HT or by an increase of the fictive temperature, the relaxation of densified silica occurs under 2.5 MeV electrons. The density decrease depends on the irradiation dose and applied temperature during HP (e.g. higher HP temperature lower relaxation). As a result of irradiation, we observed an unusual intensity increase of D1 and D2 Raman bands whereas there is a strong decrease of glass density in 350 and 440°C treated HP samples. This indicates that bond breaking mechanism leads to the formation of 3 and 4 membered rings at the expense of higher membered rings as shown by MD calculation under laser irradiation [2], which does not systematically traduce an increase of the silica density. This depends on the initial structure of the glass and rings statistic.

Kapoor S et al. Front. Mater. 4 (2017) 1.N. Shchleblanov, M. Povarnitsyn EPL 114 (2016) 26004.

Keywords: structure, density, relaxation, irradiation, silica, phosphate

^{*}Speaker