Electron irradiation effects on the activation energy distribution in densified silica

Shchedrina N.[1,2], Ollier N.[2], Mobasher M.[2], Lancry M.[1]

[1] Institut de Chimie Moléculaire et des Matériaux d'Orsay, Université Paris-Saclay, Rue du doyen Georges Poitou, 91405 Orsay cedex, France
[2] Laboratoire des Solides Irradiés, École polytechnique-CEA-CNRS, 91128 Palaiseau Cedex, France

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Densified silica glass garners significant interest owing to its enhanced mechanical strength and distinct optical characteristics, such as an increased refractive index and decreased absorption, making it ideal material for harsh environments. Various methods can induce densification in silica glass, including thermal processing, the application of high pressure, and exposure to irradiation sources like electron beams, gamma rays, X-rays, and laser emissions [1,2]. Silica glass exhibits polyamorphism, a phenomenon that refers to silica's ability to exist in multiple amorphous states with differing short-range order structures and densities. For example, the coexistence and reversible transformations between low-density amorphous (LDA) and high-density amorphous (HDA) phases have been reported and have garnered interest, although their structure and physicochemical properties are not yet fully studied. Among the densified silica phases, the silica so-called "metamict-like" phase obtained after electron irradiation at 11 GGy is of particular interest [2,3].

In this work, we analyzed a series of silica glass samples, including a high-pressure, high-temperature (HPHT) treated samples (obtained at 5GPa 1000°C) exposed to high-energy electron irradiation of 2.5 MeV at doses of 0, 10^7 Gy, and 11 GGy. Furthermore, we compared 2 metamict-like samples produced under two conditions: 11 GGy irradiation with and without HPHT treatment before, which result in the same final "state" in terms of Raman spectra and density [3]. The electron irradiation was performed using the SIRIUS facility (LSI/CEA/Ecole Polytechnique) with a current not exceeding 25 μA and a temperature not exceeding 60°C. By focusing particularly on the D_2 Raman band linked to 3-membered rings, we monitored the structural evolutions of the glass during isothermal annealing. Annealing was done at three temperatures (825 °C, 850 °C, and 900 °C) with specific intervals for Raman spectral recording. Using the data from isothermal annealing curves, we utilized the VAREPA framework and "master curve" methodology to assess the thermal stability of densification, estimating the activation energy distribution based on accelerated aging data and 1st order Arrhenius kinetics.

Our results show that while both metamict-like samples had comparable density, Raman signatures, and activation energy distribution widths (around 0.6 eV), the central energy for the HPHT treated 11 GGy sample was significantly elevated at 2.87 eV compared to the purely irradiated SiO2 11 GGy sample at 1.89 eV. This points to an enhanced thermal stability and an altered internal structure in metamict-like silica glass when exposed to HPHT. For the unirradiated densified and low-dose irradiated samples, a dual-mode activation energy distribution was present. This unveils the role of the HDA-LDA transition phase in the origin of the D_2 Raman band's non-monotonous behavior in low-irradiated densified samples. Electron irradiation seems to minimize the HDA state, prompting a shift to a LDA configuration [4].

REFERENCES

[1] Y. Wang, M. Cavillon, N. Ollier, B. Poumellec, and M. Lancry, Physica Status Solidi (2021).
[2] N. Ollier, M. Lancry, C. Martinet, V. Martinez, S. Le Floch, and D. Neuville, Scientific Reports **9**, (2019).
[3] I. Reghioua, M. Lancry, O. Cavani, S. L. Floch, D. R. Neuville, and N. Ollier, Applied Physics Letters **115**, (2019).
[4] N. Shchedrina, N. Ollier, M. Mobasher, and M. Lancry, J. Non Cryst. Solids **617**, 122491 (2023).