Electron irradiation effects on the activation energy distribution in densified silica

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Densified silica glass, having enhanced mechanical and unique optical properties, serves critical roles in extreme environments as a material for various applications, such as optical sensors. In this study, a set of samples were prepared and analyzed using Raman spectroscopy and activation energy calculation (1). The range includes the same high-pressure, high-temperature (HP-HT) treated sample subjected to three different irradiation doses – 0, 107 Gy, and 11 GGy. Additionally, metamict-like samples obtained under two distinct conditions – 11 GGy with and without HPHT treatment (2,3) – were compared. In this study, alterations in Raman spectra, particularly in the D2 band linked to 3-membered rings in silica's matrix, serve as indicators of structural transformations in the glass during annealing. Utilizing a 'master curve' approach (4) derived from these annealing curves, we assess the distribution of activation energy, offering a dependable method for forecasting the longevity of density properties. Samples were subjected to isothermal annealing, during which their Raman spectra were continuously monitored in real-time. The annealing was carried out at 3 temperatures between 825°C and 900°C, each for a duration of 18 hours. Our findings indicate that despite similar activation energy distribution widths (~0.6 eV) for both samples, the central energy was notably higher for the HP-HT treated 11 GGy sample (2.87 eV) than for the solely irradiated SiO2 11 GGy sample (1.89 eV). This suggests greater thermal stability and thus internal structure for densified silica glass subjected to HP-HT conditions. For the densified sample and the low-dose irradiated sample, we observe a bimodal activation energy distribution. This unveils the role of the high-density amorphous (HDA) transition phase in influencing the D2 Raman band's non-monotonous behavior in low-irradiated densified samples. Electron irradiation appears to minimize these HDA states, leading to a transition to a lower-density amorphous (LDA) structure.

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