Goldschmidt 2025 Abstract
https://doi.org/10.7185/gold2025.25666

Effect of Heavy-Ion Irradiation on Glass Dissolution Rates and Surface Alteration Layer Formation studied operando by Fluid-Cell Raman Spectroscopy

MRS. MARA IRIS LÖNARTZ[1], LASSE STAUSBERG[1], CHRISTINA TRAUTMANN[2] AND THORSTEN GEISLER[1]

[1]University of Bonn

[2]GSI Helmholtzzentrum Darmstadt and Technische Universität Darmstadt

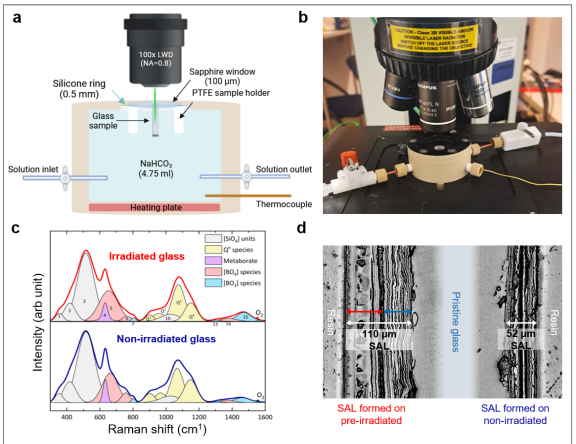

Figure 1: (a) Sketch and **(b)** photo of the Fluid-cell Raman spectroscopic setup (not to scale). **(c)** Representative background-corrected Raman spectra from the irradiated glass (upper red) and non-irradiated (lower blue) glass. **(d)** High-resolution digital microscope image of the corroded glass and the SAL formed at the pre-irradiated and non-irradiated side of the sample.

Borosilicate glass presents a preferred host material for the immobilization of high-level nuclear waste due to its compositional flexibility and corrosion resistance in aqueous environments[1,2]. In a deep geological repository, the vitrified waste will be exposed to self-irradiation damage from radionuclides and aqueous corrosion from infiltrating groundwater. However, the effect of self-irradiation damage on the structural integrity of the glass and the mechanisms and kinetics controlling the glass dissolution are poorly understood[3,4]. In order to induce severe radiation damage within a layer of approximately 50 µm, a ternary Na borosilicate glass was irradiated with 950 MeV Au ions. Real-time and *in situ* Fluid-cell Raman spectroscopic (FCRS) experiments revealed a two to threefold increase in the initial dissolution rate of the pre-damaged glass compared to non-irradiated glass, corroded in a 0.5 M $NaHCO_3$ solution and at temperatures between 80 and 85°C (Fig. 1a-c). The thickness of the irradiated layer and the associated depolymerization of the borosilicate network were found to correlate with the dissolution kinetics and textural features of the silica-based surface alteration layer (SAL). The overall thicker SAL evolved from a homogeneous to a laminar texture, revealing further changes in the effective diffusivity of molecular water through the SAL, as evidenced by D2O tracer experiments (Fig. 1d). FCRS facilitates the *operando* observation of reaction mechanisms, kinetics, and their mutual interactions with transport processes during the aqueous corrosion of borosilicate glasses, providing novel insights into the effect of irradiation damage on the glass dissolution kinetics and transport properties of the SAL.

[1] Ojovan, M. I. & Lee, W. E. (2011), *Mater. Trans. A-Phys. Metall. Mater. Sci,* 42A, 837-851.

[2] Gregg, D. J. (2025), *Bulletin of the Atomic Scientists*, 81(1), 3–16.

[3] Lönartz et al. (2019), *Materials*, *12*(9), 1480.

[4] Gin et al. (2024), *npj Mater Degrad* 8, 64.