Long-Term Occurrence and Evolution of an Accelerated Alteration Rate for Nuclear Waste Glasses in Repository Conditions

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Understanding the long-term alteration behavior of glass at geological timescales is critical to characterize the evolution of natural, archeological, as well as man-made materials such as nuclear waste glasses. Generally, glass briefly alters at a fast forward rate (Stage I) that slows to a residual rate (Stage II) that persists over long time scales. The processes that govern this long-term alteration rate are still debated. In certain conditions, an acceleration of the alteration rate is observed (Stage III). Observed for nuclear glasses at $T \geq 70°C$, Stage III is generally accompanied by the precipitation of zeolites [1]. Zeolite phases are also commonly observed in natural glass alteration down to 5°C [2].

This study focuses on 24 high Na borosilicate nuclear waste glasses statistically designed to cover a compositional range relevant to Hanford Low-Activity Waste glasses. Glasses were altered for several hundred days at four temperatures ranging from 22°C to 90°C in static testing (solution-volume to glass-surface-area ratio of 2000 m^{-1}). A small amount of zeolite Na-P2 particles were added to each experiment to initiate Stage III behavior consistently [3].

Experimental results throughout the matrix of 96 tests show different Stage III behaviors depending on the glass compostion and test temperature, ranging from a sustained accelerated rate until the glass was fully altered to a brief acceleration followed by a return to a Stage II-like behavior. A partition model fit to the experimental responses at 90°C revealed the importance of the CaO to the observation of an immediate linear response, and the importance of Na_2O mass fractions in the glass in the persistence of a sustained Stage III rate. A composition-parameter correlation model further confirmed the link between the Na_2O fraction in the glass and the magnitude of the observed Stage III rates. Temperature effect on Stage III will also be discussed.

[1] Parruzot *et al.* (2019) *J. Nucl. Mater.* **523**, 490–501. [2] Crovisier *et al.* (2003) *J. Nucl. Mater.* **321**, 91–109. [3] Fournier *et al.* (2017) *npj Mat. Degr.* **1** 1.