A New Model for Nuclear Waste Borosilicate Glass Corrosion

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Understanding the mechanism of aqueous corrosion of nuclear waste borosilicate glasses is essential to reliably predict their long-term aqueous durability in a geologic repository. Here we report the results of corrosion experiments with borosilicate glass cuboids with edge lengths of about 2.5mm in a HCl solution of pH = 0 at 150°C for 6 to 336 hours, including two experiments with ^{18}O and ^{26}Mg as isotope tracers. Several observations were made in this study such as (i) the occurrence of chemical oscillations in the corrosion rim that is composed of silica, (ii) an enrichment of ^{18}O and ^{26}Mg in the corrosion rim formed in the isotopically enriched solutions without observable diffusion profiles, (iii) a sharp phase boundary of the corrosion rim towards the pristine glass, (iv) a high porosity in the corrosion rim, and (v) the occurrence of silica spherules at the surface. These features are not at all compatible with classical theories about the formation of the corrosion or "gel" layer that are based on diffusion-controlled hydration and ion exchange reactions and subsequent solid-state re-condensation of the hydrolyzed glass network (Grambow 2006, and references therein). We propose a new mechanistic model for glass corrosion that is based on congruent dissolution of the glass that is spatially coupled to the precipitation of amorphous silica at an inward moving reaction front. The model provides a novel framework to evaluate the long-term performance of nuclear waste glasses.

References

Grambow B. (2006) Nuclear Waste Glasses - How Durable? Elements 2: 357-364.

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