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Surface Layer-Bulk Glass Interface Evolution with Aqueous Corrosion

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ABSTRACT

Most nations have turned to the vitrification of high-level wastes (HLW) for disposal of waste generated through power generation and weapons manufacture. In fact, in the U.S., the Environmental Protection Agency (EPA) has declared vitrification the best demonstrated available technology for HLW stabilization. Therefore, the ability to predict long-term (100,000 years+) HLW glass corrosion performance, and hence radionuclide release rates, is vital. However, this remains a challenging task. It has been proposed that three potential mechanisms could be controlling release rates of radionuclides from the glass at long time periods: affinity control, transport control, and ion exchange. Prior to the development of a defensible, consensus rate law, the rate limiting mechanism for radionuclide release must be determined. It has been proposed that processes at the surface layer-bulk glass interface are the rate limiting steps in the long term aqueous corrosion of glasses. However, little is known about the properties of, or the processes that occur at, this interface.

The overall objective of this proposal is to investigate the morphology, structure and chemistry of the surface layer-bulk glass interface and its impact on the long-term reaction kinetics of borosilicate-based HLW glasses. We hypothesize the bulk glass surface morphology and structural transformations that occur at the surface layer-bulk glass interface throughout aqueous corrosion will control the overall mechanism and dissolution behavior of borosilicate based HLW glasses. This hypothesis will be tested through a series of *in-situ* and *ex-situ* experiments under experimentally simulated long-term reaction conditions. The bulk glass surface morphology and structure at that interface will be examined through *in-situ/ex-situ* atomic force microscopy and x-ray scattering techniques while its chemistry will be analyzed through *ex-situ* spectroscopic techniques. Long-term reaction conditions of the surface layerbulk glass interface will be reproduced through systematic changes in glass composition/structure, glass surface morphology, and reactive solution chemistry/solubility. In particular, it is hypothesized that the solution composition at the interface is different than that in the bulk solution and this difference may be a driving force in the mechanism of glass corrosion at long time periods. We will test this hypothesis specifically, through modeling of the interfacial solution pH and solubility with respect to glass components, followed by experimentation under modeled conditions.