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Formation of Zeolites Responsible for Waste Glass Rate Acceleration: An Experimental and Computational Study for Understanding Thermodynamic and Kinetic Processes

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Program: FC-1.3b, Waste-Form Development: Zeolite Formation Thermodynamics and Kinetics
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ABSTRACT:

Before nuclear waste glasses can be disposed at geological disposal facilities, a performance assessment must be conducted to provide confidence to the regulators and public that the release rates of radioisotopes and hazardous materials into the environment are below the regulatory limits. The release rate of radionuclides is directly related to the dissolution rate of the radionuclide-containing glass; therefore, the corrosion behavior of the glass must be well understood. In static conditions, glass dissolution is categorized into three different stages where the least understood stage (Stage III) has the possibility to drive radionuclide release rates above regulatory thresholds. While the onset of Stage III glass dissolution cannot be predicted, it is related to the formation of a zeolitic phase(s). The overall goal of this project is to both understand the formation of zeolite phases and to develop methods to control processes thereof in order to suppress Stage III dissolution.

This project will leverage unique expertise in zeolite chemistry (lead institution), glass corrosion (national lab), and modeling zeolite formation (university partner) to examine zeolite formation and methods to impede their growth to prevent the advent of Stage III behavior. This proposal is particularly innovative in its approach to deliver thermodynamic and kinetic information that can be directly implemented in performance assessment calculations. The rationale for this work is to better understand the degradation processes for waste forms that could be generated in advanced nuclear fuel cycles, which will enable increased reliance on waste form performance in repository environments. To this end, the following three objectives will be addressed in this project through the synergistic combination of experimental and computation studies:

Objective 1: Examine the kinetics of zeolite formation at low temperatures relevant to geological repositories. Experimental efforts will be coupled with Density Functional Theory (DFT) modeling of the mechanism and energetics of zeolite formation. We posit that changes in temperature and pH will lead to notable reductions in the rate of zeolite growth and predominant zeolite phase, and potentially impede its formation or direct the growth of crystal polymorphs that impact the adverse effects of Stage III behavior.

Objective 2: Investigate glass dissolution in the presence of zeolite seeds at low temperatures and over a range of pH using a variety of glass compositions to understand the effect of glass and zeolite composition on the rate and probability of Stage III resumption.

Objective 3: Identify new methods to impede Stage III glass dissolution via the introduction of alkali and alkaline earth metals that can either function as structure breakers that reduce the rate of zeolite growth, or as structure-directing agents that promote the formation of different zeolites. We anticipate these findings will aid in the design of new glass formulations and the selection of geological repository sites.