Molecular Dynamics-based Simulations of Bulk/Interfacial Structures and Diffusion Behaviors in Nuclear Waste Glasses

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ABSTRACT:

We aim to generate accurate atomic structural models of nuclear waste glasses by using large-scale molecular dynamics-based computer simulations and to use these models to investigate self-diffusion behaviors, interfacial structures, and hydrated gel structures formed during dissolution of these glasses. The goal is to obtain realistic and accurate short and medium range structures of these complex oxide glasses, to provide a mechanistic understanding of the dissolution behaviors, and to generate reliable information with predictive power in designing nuclear waste glasses for long-term geological storage.

Vitrification is an effective way to immobilize high and medium level nuclear wastes. A detailed understanding of the structure and properties of these glasses is essential in elucidating the dissolution behavior and longer term chemical durability of these glasses. Unlike crystalline materials, the structure of these multicomponent oxide glasses lacks long range order and their structures cannot be uniquely determined by any experimental methods. In this project, we use atomistic computer simulations to generate realistic structure models of nuclear waste glasses. Careful testing of potential models and validation of the simulated glass structures with experimental characterizations will be performed. The generated glass structures will be further used to study the ionic diffusion in the glasses and studying the glass/water interfaces and the dissolution gel structures. In addition, the results of this project will provide realistic structures for input in Monte Carlo simulations of the full dissolution process, a method that has shown great potential in understanding dissolution of glasses in realistic time scale but is currently limited by the use of idealized glass structure models based on simple lattices.

Our research team has expertise in several key areas of this project. The PI, Dr. Jincheng Du, has extensive experience in modeling glass structure and properties and in potential development. His work has been funded by National Science Foundation (NSF) including one project on dissolution behaviors of bioactive glasses and one NSF GOALI project on mixed glass former effect on industrially important glass systems with Corning Inc. He currently chairs the Technical Committee on atomistic modeling and simulations of glasses of International Commission of Glass (ICG). Co-PI Dr. Louise J. Criscenti has led several projects on the study of the simulations of the structure and behaviors of glass waste forms in geological environments. Co-PI Dr. Sebastien Kerisit has extensive experience in atomistic modeling of mineral and mineral/water interfaces. He has also developed and implemented a Monte Carlo code for simulating the corrosion of borosilicate glasses. Co-PI Dr. Joe V. Ryan provides expertise of experimental characterization of the waste form glasses and deep understanding of experimental world of dissolution behaviors of glasses. About 20% of the requested fund goes to national labs collaborators.

Educating next generation researchers and engineers with experiences on nuclear energy and nuclear materials is another important aspect of this project. The diverse background of this project will provide student and postdoc researchers a great opportunity to work on the scientifically challenging problems on nuclear waste glasses and their chemical durability. They will visit and be trained at the national lab facilities as summer interns and work closely with national lab collaborators.