

Thermochemical Modeling of High-Level Waste Glass and Application To Waste Form Stability

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Surface Processing & Mechanics



Objective of Research

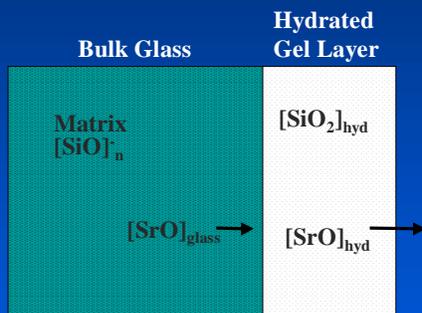
Develop practical mode for calculating thermodynamic stabilities of components in complex high level waste glass and spent nuclear fuel.

- ❖ Work with large number of components
- ❖ Easy to understand and use
- ❖ Reliable
- ❖ Extrapolatable and interpolatable (T, x_i)

Relevance to EMSP

- ❖ Work will provide a thermochemical model for waste glass and spent nuclear fuel
- ❖ Model allows prediction of liquidus temperatures and precipitation of undesirable crystalline phases
- ❖ Results provide activities for reaction paths for leaching/transport models and codes such as ESP
- ❖ Predicted activities allow anticipation of effects such as corrosion and potential for release
- ❖ Expected understanding will add confidence to the prediction of stability over long time periods
- ❖ Information can be used in designing and engineering waste forms and barriers
- ❖ The work may provide basic insights into solid solution behavior, bonding in glasses, and crystal chemistry of fluorite structure oxides

Example of Potential Improvement to Predicted Leaching Rates in Waste Glass Yields 5 Orders of Magnitude Difference in Chemical Activity for SrO



Liquid Leachant

The concentration of the leached waste ion is dependent on the activity of [SrO]_{glass}

	Concentration SrO in glass	Activity of [SrO] _{glass}	Concentration In Leachant
Ideal Solution Assumption	6.8 mol%	0.068	0.068
Associate Model*	6.8 mol%	1.8x10 ⁻⁸	1.8x10 ⁻⁸ m

*Aluminosilicate glass (Spear and Pantano, 1997)

Associate Species Model Is Being Used to Represent HLW Glass and Spent Fuel

- ❖ The glass thermochemistry is modeled as a solution of constituent compounds
 - e.g., Soda-alumina glass modeled as ideal solution of Na₂O + AlNaO₂ + Al₂O₃
 - even though there are at least two more phases in the system
- ❖ Great simplification over previous methodologies
 - Treat glass thermochemically as a supercooled liquid
 - No extra parameters that multiply as systems become complex
 - Parameters inherent in species selected for inclusion
 - Allows further simplification by eliminating species that are not "important"

