Unraveling the Complex Solution Chemistry of Aluminum in Hanford Site Nuclear Tank Waste

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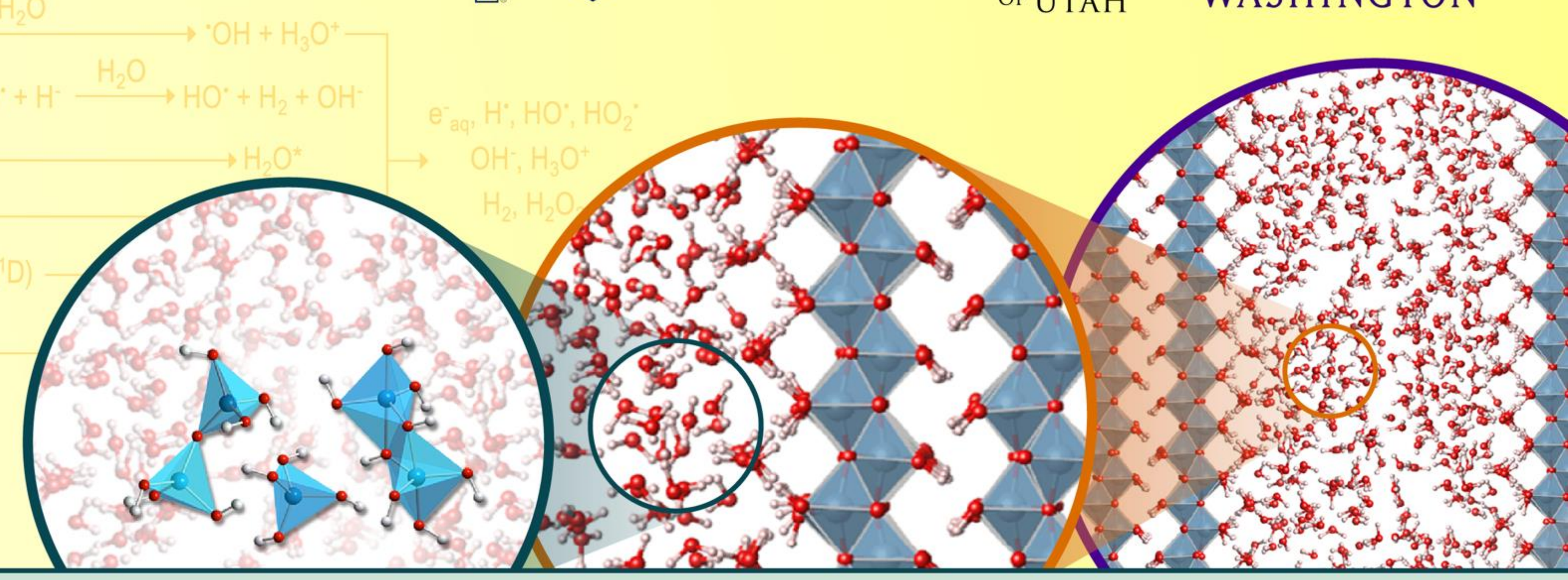

Overview

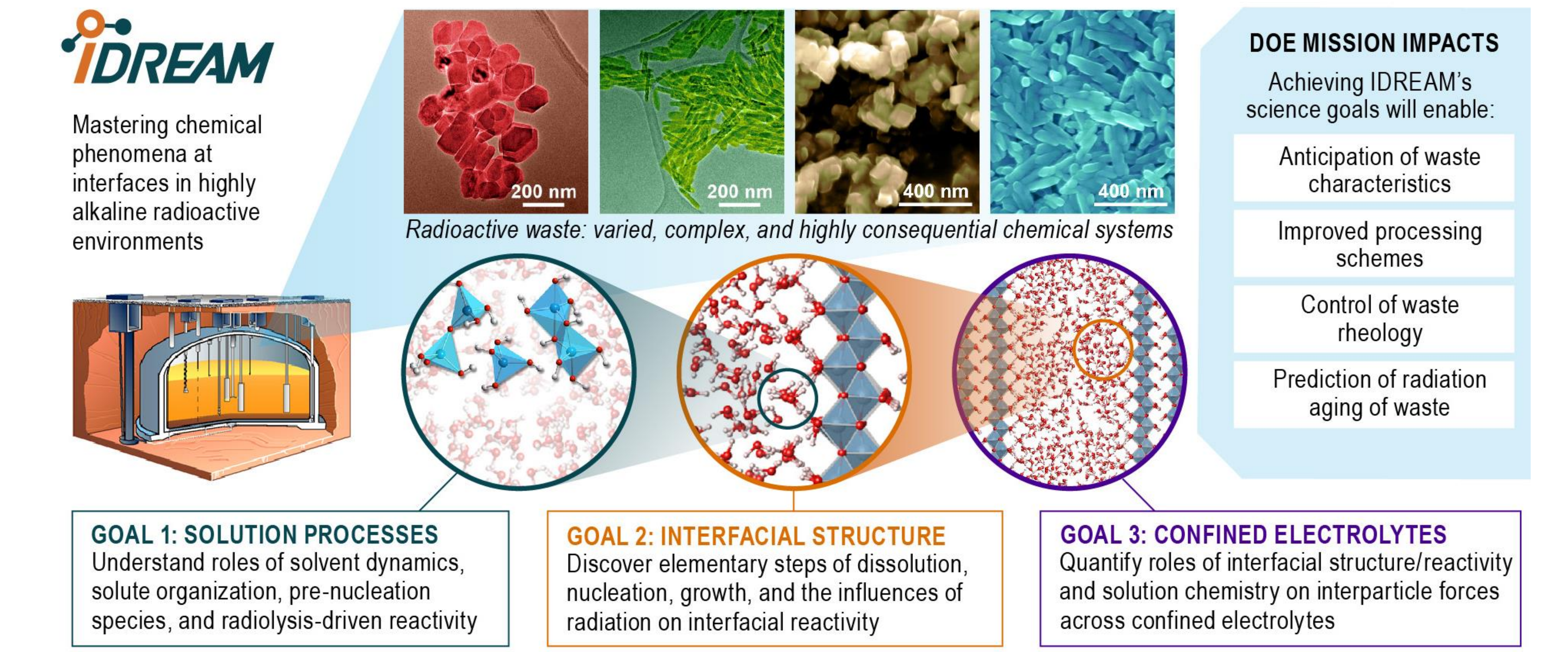

Figure 1. IDREAM is structured to provide the fundamental science basis to speed up processing of the millions of gallons of highly radioactive wastes stored at DOE's Hanford and Savannah River Sites.[2]

Introduction

- ~56 million gallons of radioactive waste divided between 177 underground tanks is present at the Hanford Site[1]
- The Interfacial Dynamics in Radioactive Environments and Materials (IDREAM) Energy Frontier Research Center seeks to understand the complex chemical environments present in nuclear waste (Figure 1)
- The solubility, nucleation, crystallization, and aggregation behavior of aluminum is of interest
- Al is present in nuclear tank waste as octahedrally coordinated aluminum hydroxide polymorphs which crystalize in an unpredictable manner (Figure 2)
- Previously, the solubility of gibbsite ($Al(OH)_3$) was seen to increase in deuterated and nitrate/nitrite rich systems[3,4]
- In this work, the dissolution kinetics of gibbsite will be determined under normal and deuterated conditions at various temperatures to better understand the connection between gibbsite solubility and proton transfer

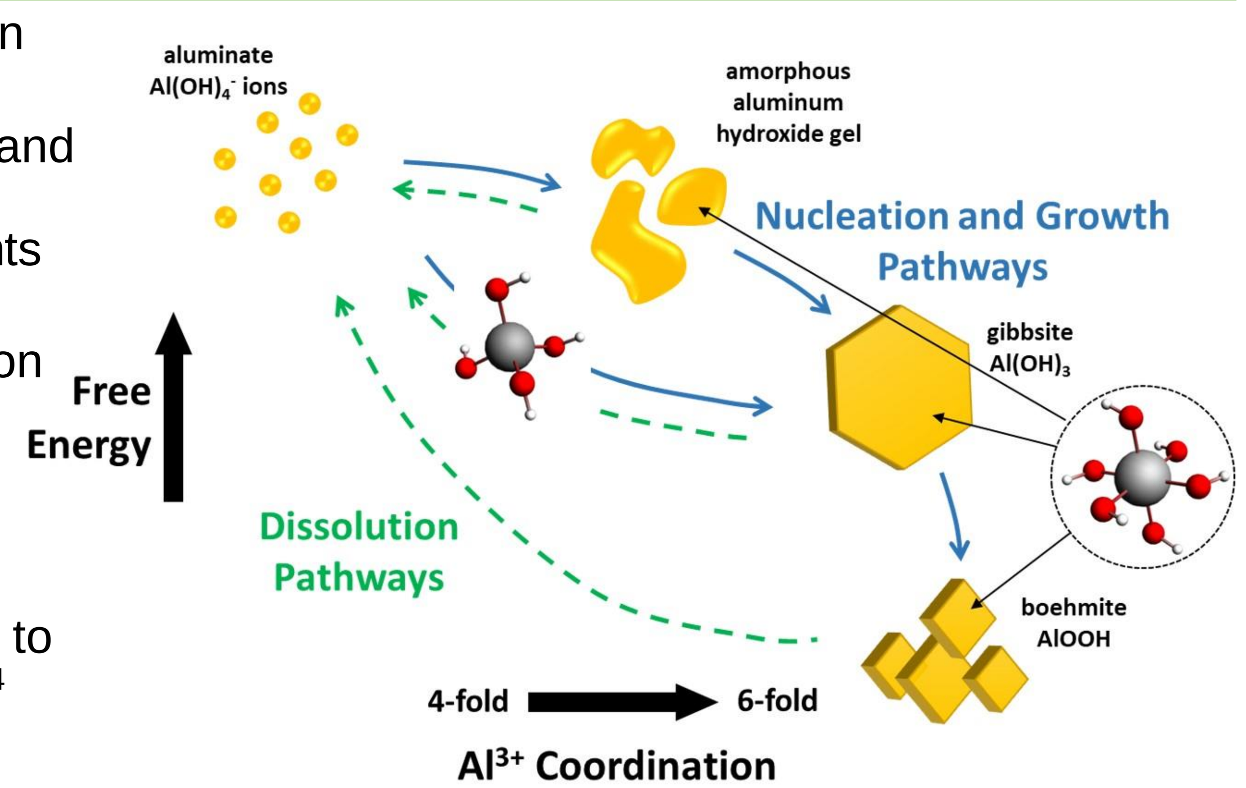

Figure 2. Diagram of the dissolution and nucleation pathways of gibbsite.

References

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Previous Results

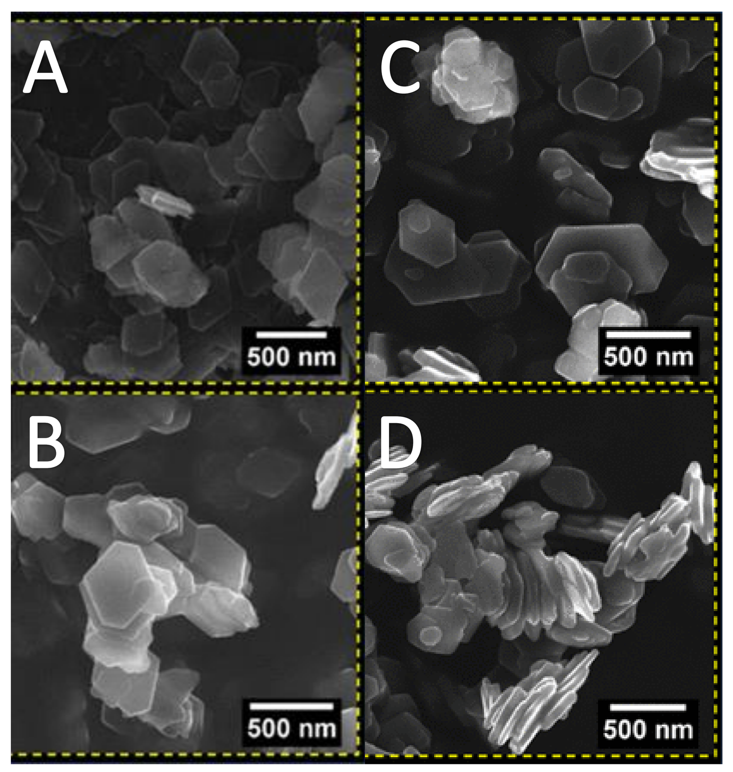

Figure 3. SEM images of gibbsite precursor (A) reacted in NaOH (B), NaOH/D (C), NaOD (D).[3] Solution structure effects gibbsite dissolution and reprecipitation.

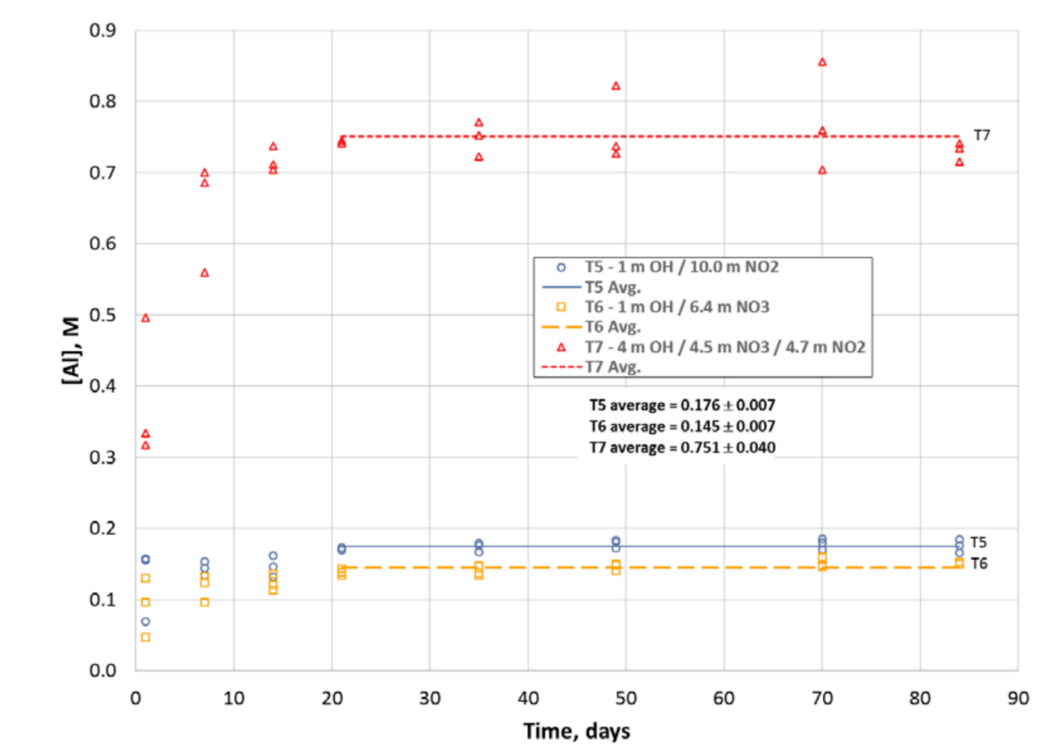

Figure 4. Gibbsite solubility increases in a mixed nitrate/nitrite solution vs a non-mixed solution.[4]

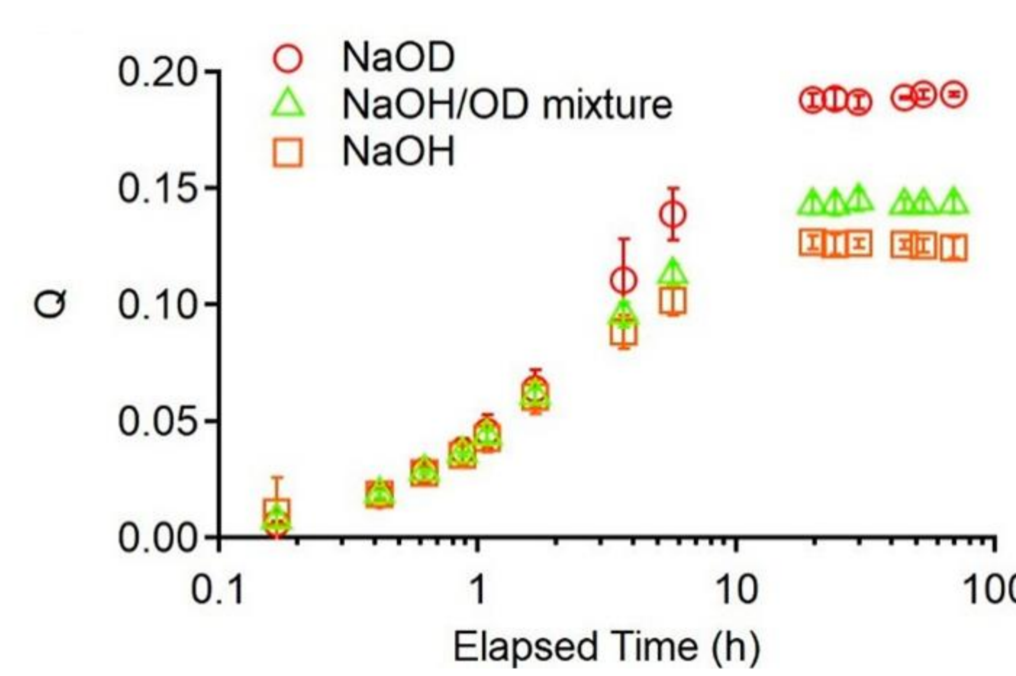

Figure 5. ICP-OES data depicting gibbsite solubility increasing in a deuterated system.[3]

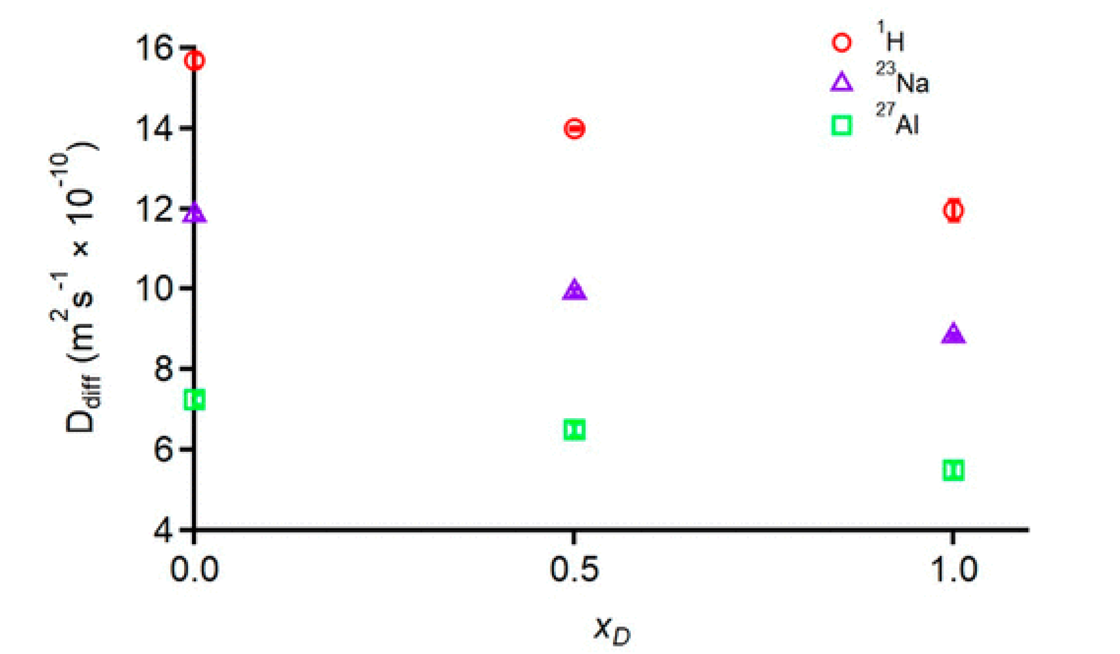

Figure 6. NMR data showing the decreasing diffusion coefficients of gibbsite as a function of the total ratio of deuterium.[3]

Discussion & Future Directions

Previous works left unanswered questions, including how the kinetic effects of deuterium-substitution alters gibbsite diffusion and crystallization in complex alkaline conditions. This project proposal looks to determine whether gibbsite solubility changes with temperature or if the dissolution changes are due to the kinetic effects of a shifting H-bonding network. One of the goals of IDREAM is to determine the mechanisms required for a predictive understanding of aluminum hydroxide crystallization in Hanford tank waste. This project will build upon previous work to gain a better understanding of how deuterium, a product of radiation, effects the precipitation of gibbsite in tank waste. Moving forward, this project will alter previous methodologies as follows: A) use of deuterated gibbsite, B) reduced reaction time, C) additional analysis techniques, D) use of four temperature points: 20, 40, 60, 80 °C.

Acknowledgements

This research was supported by IDREAM (Interfacial Dynamics in Radioactive Environments and Materials), an Energy Frontier Research Center funded by the U.S. Department of Energy (DOE), Office of Science, Basic Energy Science (FWP 68932). PNNL is a multiprogram national laboratory operated for DOE by Battelle Memorial Institute operating under Contract No. DE AC05-76RL0-1830. This work was also supported by the Department of Energy Office of Environmental Management – Minority Serving Institutions Partnership Program (EM – MSIPP).