Poster Title: **New NMR Methods for Studying the Impact of Mineral Transformation and Sorbate Aging on Contaminant Speciation and Mobility**

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The work of our collaborative research team is aimed at developing a predictive-mechanistic understanding of the coupling between mineral weathering from caustic waste release and contaminant (Cs, Sr, I) fate and transport in waste-impacted sediments across space, time and geochemical gradients that encompass the process-level heterogeneity observed at the Hanford DOE site. In these studies we have examined sets of samples ranging from model clay minerals to Hanford sediment samples, each reacted with a simulated tank waste leachate (STWL) containing Sr, Cs, and/or I in batch laboratory experiments from 1 d to 1 yr.

One of our tools in this research is solid-state nuclear magnetic resonance (NMR). Preliminary studies focused on the identification and quantification of neophases formed in a kaolinite/STWL system as a function of reaction time, where variable-field ^{27}Al magic-angle spinning (MAS) NMR provides kinetic data for the formation of four aluminum-containing neophases. Additional studies on other model clays have also progressed, and new information from data acquired at high magnetic field strengths will be presented and discussed. These data have been acquired at the Environmental Molecular Sciences Laboratory, a national scientific user facility sponsored by the Department of Energy's Office of Biological and Environmental Research and located at Pacific Northwest National Laboratory (PNNL). Additional information regarding the coordination and identification of aluminum atoms in neophase systems has also been accessed via multiple-quantum magic-angle spinning (MQMAS) NMR experiments, and implementation of these methods will be demonstrated in studies of cesium siting in aluminum-containing phases.

Solid-state NMR studies of ^{87}Sr can provide details of the local environments of the Sr atoms and hence their location in complex reaction product mixtures. However, ^{87}Sr is a very difficult nucleus to detect via NMR, and therefore we have explored means to enhance sensitivity by performing quadrupolar Carr-Purcell Meiboom-Gill (QCPMG) experiments using the 21.14 T NMR spectrometer at PNNL. Improvements in signal-to-noise gained by using the QCPMG sequence at high field result in up to ten thousand-fold reductions in experiment time, and we will discuss our ^{87}Sr NMR results in mineral systems as well as the feasibility of detecting Sr resonances in clays, zeolites, and feldspathoid systems.

Finally, we report progress toward the measurement of reactive surface area on complex oxides in the environment. Probe molecules containing fluorine are selectively attached to reactive sites on the surfaces of oxide materials, and quantitative ^{19}F MAS NMR measurements have demonstrated linear scaling of reactive surface sites with surface dissolution rates. Our measurements on aluminosilicate glasses and clay materials will be presented, and possible applications of these methods to more complex sediment samples (both pristine and reacted) will be discussed.

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