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The Interaction of Strontium with Kaolinite under Extreme Environmental Conditions: An EXAFS Study

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Introduction: Sorptive interactions with various natural soil particles play an important role in controlling the speciation and, hence, the mobility of radionuclides, such as Sr^{2+} in the vadose zone. Geochemical changes caused by the introduction of corrosive wastes at some DOE sites (e.g. Hanford, WA; Savannah River Site, Aiken, SC) affect the surficial properties of soil particles, but how such changes influence the binding and retention of metals are not known. A number of previous studies focused on dissolution and precipitation rates of clay systems in acidic to neutral conditions without the presence of radionuclides. However, there is lack of information concerning sorption, dissolution and precipitation kinetics of clays at high pH and ionic strength with radionuclides present, which is representative of the actual conditions in many sites. In this study, we used EXAFS to investigate the molecular environment of Sr sorbed on kaolinite reacted with synthetic tank waste leachate (STWL).

Methods and Materials: The composition of the STWL was 2 M NaNO_3 solution, pH 14, and 0.05 M $\text{Al}(\text{OH})_4^-$. Samples of Sr adsorbed on kaolinite were generated by reacting kaolinite in a STWL medium for the time periods 7, 90 and 180 days, in the presence of 10-3 M Sr. The residue was separated by centrifugation, washed and dried for EXAFS analysis.

Results: Figure 1 shows the Fourier Transforms of the EXAFS spectra representing radial structure function for Sr-sorbed on kaolinite as a function of time in the STWL medium. The major peak at an uncorrected distance of 2.07 D represents an oxygen neighbor which lies at a bond distance of ca. 2.56 D in SrCO_3 according to earlier work [1]. Our results indicate the presence of two other scatterers surrounding Sr, which, after applying a correction factor of 0.5 D, occur at a distance of ca. 3.47 D and 4.34 D. The scatterer at 4.34 D is likely to be a Sr atom. However, this peak disappears in samples reacted with STWL for longer than a month with the concomitant formation of a new peak at a corrected radial distance of 3.47 D. These results clearly suggest that kaolinite is significantly altered when reacted with STWL, thereby strongly influencing its interaction with Sr. X-ray diffraction studies suggest that kaolinite transforms with time to zeolite structures, resembling chabazite, sodalite and cancrinite. Thus, it is possible that Sr can become incorporated into such zeolite structures. Consequently, the new peak formed at at 3.47 D may represent an Al atom in the neighborhood of Sr incorporated into a zeolite structure.

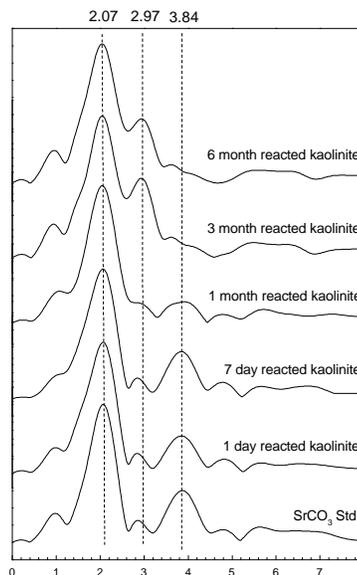


Figure 1. Fourier Transforms showing radial structure function for Sr sorbed on kaolinite as a function of time in the STWL medium.

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References: (1) R.H. Parkman, J.M. Charnock, F.R. Livens and D.J. Vaughan, "A Study of the Interaction of Strontium ions in Aqueous Solution with the Surfaces of Calcite and Kaolinite," *Geochim. Cosmochim. Acta*, **62** (9), p. 1481-1492, 1998.