Goldschmidt 2000
September 3rd–8th, 2000
Oxford, UK.

Journal of Conference Abstracts
Volume 5(2), 1070

Extreme Geochemistry: Strontium Interactions in Hyper-alkaline Aluminum and Feldspar Systems

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High concentrations of aluminium exist in the radioactive waste held in leaking tanks at the United States Department of Energy's Hanford, Washington site. The aluminum originated from chemical processing techniques applied to the waste, which is also the source for the high concentrations of sodium hydroxide responsible for the extreme alkalinity of the waste. The waste is held in large subsurface tanks, many of which are leaking into the surrounding groundwater and soil. There is concern about the fate of these leaking solutions and their interactions with the surrounding geomedia.

One possibility is that these caustic solutions will promote the formation of secondary mineral phases through dissolution of existing minerals and that radioisotopes, present in the leaking tank waste, may be incorporated into the newly formed solids.

This study focuses on the nature of the aluminum species found in high aluminum concentration and high pH solutions, the dissolution characteristics of potassium feldspar in the presence of these alkaline aluminate solutions and how these conditions impact strontium sorption. Synchrotron-based spectroscopic techniques are used to elucidate the chemical speciation at the solid-solution interface.

Previous work investigating aluminum species in alkaline solution has used ^{27}Al NMR spectroscopy but Al K-edge (x-ray absorption spectroscopy) XAFS provides unique, element-specific information on the chemical form and speciation of Al surface complexes and precipitates. This technique was used to characterize the aluminum species that exist at the solid surface under high pH conditions. In addition, x-ray photoelectron spectroscopy (XPS), scanning electron microscopy (SEM) and Al K-edge XAFS were used to characterize changes to the potassium feldspar surface. This aspect of our study complements previous work on the acid dissolution behavior of feldspar (Casey et al., 1988). Strontium K-edge extended x-ray absorption spectroscopy (EXAFS) was used to identify the local chemical structure of the predominant Sr interfacial species in the feldspar and feldspar-aluminum systems.

Macroscopic and molecular-scale spectroscopic studies on strontium sorption showed significant differences between strontium sorption behavior in different systems. Variations in pH had only a minor impact on the extent of Sr uptake in systems without added aluminum. However, strontium uptake increased dramatically in feldspar systems after the addition of aluminum and variations in uptake with pH were larger.

The present work presents new fundamental geochemical information relevant to strontium contamination and mobility in hyper-alkaline environments. These findings increase our understanding and ability to predict the complex geochemical processes occurring beneath caustic waste storage areas associated with the nuclear industry.

Casey WH, Westrich, HR & Arnold GW, *Geochim. Cosmochim. Acta*, **52**, 2795-2807, (1988).