Mineralogical Characteristics of Yucca Mountain Alluvium and Effects on Neptunium(V) Sorption

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Saturated alluvium is expected to serve as an important natural barrier to radionuclide transport at Yucca Mountain, the proposed geological repository for disposal of high-level nuclear wastes. $^{237}Np(V)$ (half-life=$2.4*10^5$ years) has been identified as one of the radionuclides that could potentially contribute the greatest dose to humans because of its relatively high solubility and weak adsorption to volcanic tuffs under oxidizing conditions. Our previous studies suggested that the mineralogical characteristics of the alluvium play an important role in the interaction between Np(V) and the alluvium. The purpose of this study is to further evaluate the mineralogical basis for Neptunium (V) sorpion by saturated alluvium located down-gradient of Yucca Mountain.

Quantitative X-ray Diffraction (QXRD), Scanning Electron Microscopy (SEM)/Electron Probe Microanalysis (EPMA), and Surface Area Analysis (BET) were used to characterize the mineralogy of the alluvium samples. Batch adsorption experiments were carried out to examine the effects of minerals present in the alluvium on sorption behavior of $^{237}Np(V)$ under ambient temperature and pressures. $^{237}Np(V)$ in solution was analyzed by Liquid Scintillation Counting. The carbonate content in the alluvium was determined by an acetic acid dissolution method. The iron content in the alluvium was determined by a citrate-dithionite extraction method.

The QXRD results show that the dominant phases in the alluvium are quartz, feldspar, and cristobalite followed by smectite and clinoptilolite. The results also show that the smectite and clinoptilolite fraction in the alluvium depends on the particle size range of the sample, with a larger percentage of both minerals being associated with smaller sizes. The particle sizes smaller than 0.25 μm (diameter) consist of more than 96 weight percent of clay minerals. The sorption capacity of these clay minerals for Np(V) is about 100 times larger relative to the bulk alluvium which typically contains < 10 wt% clay minerals. These results suggest that the fine clay fraction in Yucca Mountain alluvium plays an important role in radionuclide transport near Yucca Mountain, both from the standpoint of attenuating radionuclides from solution, and of facilitating the transport of radionuclides that are strongly sorbed to clay minerals which may become mobilized as colloids. QXRD results also indicate that calcite occurs as a relatively minor phase in the alluvium samples. The lack of a QXRD determination of iron oxide suggests that the iron determined by wet chemistry and EPMA must occur as either an amorphous iron oxide coating on the aluminosilicates or as a partial replacement of aluminum by iron in the octahedral layer of smectite. Our batch experiments indicate that, in general, the Np(V) K_d values are correlated with particle size, surface area, and the total amount of smectite and clinoptilolite.