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Sorption mechanisms of Sr and Pb on zeolitized tuffs from the Nevada test site as a function of pH and ionic strength

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ABSTRACT

The sorption of Sr^{2+} and Pb^{2+} on zeolitized tuffs from the Nevada Test Site (NTS) was investigated using macroscopic batch sorption experiments and X-ray absorption spectroscopy (XAS) as a function of geochemical parameters, including pH, ionic strength, and type of background electrolyte. The sorption of Sr^{2+} is dependent on the ionic strength of the medium and independent of pH, suggesting that Sr^{2+} sorption is controlled by ion exchange at permanent charge sites. At higher ionic strengths, background electrolyte cations compete effectively with Sr^{2+} for cation-exchange sites and Sr^{2+} sorption is suppressed. At the two lower ionic strengths (0.01 and 0.1 *M*), Pb^{2+} sorption is also consistent with adsorption by cation exchange. At the highest ionic strength (1.0 *M*), however, exclusion of Pb^{2+} from cation-exchange sites resulted in pH dependent adsorption, consistent with sorption on amphoteric surface hydroxyl sites or formation of surface precipitates. XAS was used to test these hypotheses. Based on XAS data, Sr^{2+} formed hydrated surface complexes coordinated with approximately eight O atoms at an average distance of 2.60 (±0.02) Å, regardless of conditions, consistent with the formation of mononuclear, outer-sphere surface complexes at the Ca2 site in the B channel of clinoptilolite. The coordination environment of sorbed Pb^{2+} was more complex and a function of pH and ionic strength. The first shell consisted of two to three O atoms at an average distance of 2.20 (±0.02) Å. At low pH and ionic strength, XAS data were consistent with Pb^{2+} adsorption at the Na1 and Ca2 cation exchange sites in channels A and B of clinoptilolite, respectively. At the highest ionic strength (1.0 *M*) and low pH, XAS provides evidence for formation of Pb^{2+} monodentate, corner-sharing inner-sphere complexes, whereas at higher pH, XAS analysis is consistent with formation of edge-sharing bidentate inner-sphere complexes. As surface coverage increased, appearance of a second Pb^{2+} peak suggests the formation of polynuclear, inner-sphere surface complexes. These results have significant implications for the transport of radionuclides and other contaminants at the NTS and other nuclear test sites and for the modeling of these processes.