

Alteration, adsorption and nucleation processes on clay-water interfaces: mechanisms for the retention of uranium by altered clay surfaces on the nanometer scale

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Nano-scale processes on the solid-water interface of clay minerals play an important role in controlling the mobility of metals in the environment. These processes can occur in confined pore spaces of clay buffers and barriers as well as in contaminated sediments and involve a combination of alteration, adsorption and nucleation processes of multiple species and phases. This study characterizes nano-scale processes on the solid-water interface between clay minerals and uranyl-bearing solution of near neutral pH from a U mill and mine tailings at Key Lake, Saskatchewan, Canada. Alteration zones around the clay minerals montmorillonite and kaolinite are characterized by different generations of secondary silicates containing variable proportions of adsorbed uranyl- and arsenate-species and by the intergrowth of silicates with the uranyl-minerals. Adsorption of U and nucleation of uranyl minerals are strongly controlled by the presence of the adsorbed oxy-anion species arsenate and silica on the reactive solid-water interface of Fe-enriched silicates. Heterogeneous nucleation of nano-crystals of the uranyl minerals occurs most likely on adsorption sites of binary uranyl-, arsenate- and silica complexes as well as on ternary uranyl-arsenate or uranyl-silicate complexes. The uranyl minerals occur as aggregates of misoriented nano-size crystals and are the result of supersaturated solutions and a high number of nucleation sites that prevent the formation of larger crystals through Oswald ripening. The results of this study provide an understanding of interfacial nano-scale processes between uranyl species and the reactive surfaces of altered clay minerals.