

Experimental Investigation of Uranium(VI) Adsorption onto Montmorillonite at Elevated Temperatures

BEN URICK¹, RUTH TINNACHER¹ AND CHRISTOPHE TOURNASSAT^{2,3}

¹California State University East Bay

²ISTO, Université d'Orléans

³Earth and Environmental Sciences Area, Lawrence Berkeley National Laboratory, Berkeley, California

Presenting Author: burick@horizon.csueastbay.edu

Uranium (U) is the primary element in spent nuclear fuel, and from an environmental perspective a potential contaminant of water resources. In future nuclear waste repositories, the potential release of U(VI) into the natural environment will be strongly limited by its adsorption onto bentonite buffer materials in engineered barrier systems (EBS), surrounding nuclear waste packages. More specifically, U(VI) adsorption onto montmorillonite clay is expected to control contaminant retardation and diffusive fluxes across the EBS, and determine the concentrations and chemical solution speciation of U(VI) fractions remaining in pore water solutions. At the beginning of the storage period, the thermal loading of waste packages and release of decay heat will create elevated temperatures in the EBS. These elevated temperatures could potentially affect U(VI) mobility due to temperature effects on a variety of parameters and processes, such as mineral transformations and subsequent changes in mineral surface characteristics, or changes in U(VI) solution speciation, adsorption and diffusion characteristics. Previous studies have shown an increase in U(VI) adsorption/precipitation for Fe(III)-saturated montmorillonite [1], and comparable or slightly increased U(VI) adsorption for illite [2], when experiments were performed at elevated temperatures. In contrast, a decrease in U(VI) adsorption onto previously heat-treated bentonite was observed when adsorption experiments were conducted at room-temperature [3]. The goal of this study is to characterize U(VI) adsorption onto natural montmorillonite as a function of pH, and over a range of elevated temperatures, in bench-scale experiments.

In this presentation, we will provide a detailed description of our experimental setup, and discuss the inherent challenges involved with temperature-dependent U(VI) adsorption experiments. Furthermore, we will compare our first experimental results with existing adsorption data from the literature and our own model predictions of U(VI) adsorption onto montmorillonite at elevated temperatures (for details on modeling, see companion presentation by Tinnacher et al.).

[1] Lu et al., *Radiochim. Cosmochim. Acta*, 2016.

[2] Maia, F., *Doctoral Thesis*, IMT Atlantique – Comue University of Bretagne Loire, 2018.

[3] Fox et al., *Appl. Geochem.*, 2019.

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