



# Uranium and Cesium sorption to bentonite colloids under carbonate-rich environments: Implications for radionuclide transport

Emily L. Tran<sup>a</sup>, Nadya Teutsch<sup>b</sup>, Ofra Klein-BenDavid<sup>c,d</sup>, Noam Weisbrod<sup>a,\*</sup>

<sup>a</sup> The Zuckerman Institute for Water Research, The Jacob Blaustein Institutes for Desert Studies, Ben Gurion University of the Negev, Midreshet Ben Gurion 84990, Israel

<sup>b</sup> Geological Survey of Israel, Jerusalem 9550161, Israel

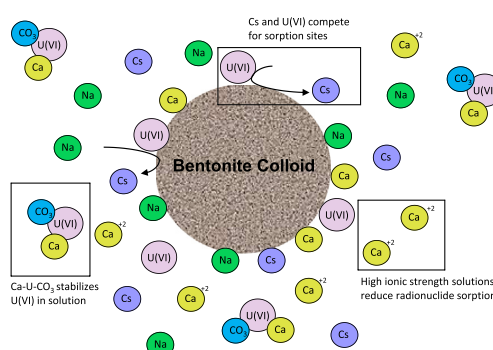
<sup>c</sup> Nuclear Research Center of the Negev, Negev, P.O. Box 9001, Beersheva 84190, Israel

<sup>d</sup> Geological and Environmental Science Department, Ben Gurion University of the Negev, Beersheva 84105, Israel

## HIGHLIGHTS

- Batch experiments determined sorption of U(VI) and Cs under environmental conditions.
- Experiments show sorption in high salinity groundwater and low salinity rainwater.
- $K_d$  for both elements increased with decreasing colloid concentration.
- Presence of U(VI) reduced Cs sorption to bentonite surfaces, and vice versa.
- Transport models should account for colloid concentration and competitive sorption.

## GRAPHICAL ABSTRACT



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## ABSTRACT

In the context of geological disposal of radioactive waste, one of the controlling mechanisms for radionuclide migration through subsurface strata is sorption to mobile colloidal bentonite particles. Such particles may erode from the repository backfill or bentonite buffer and yield measurable (0.01–0.1 g/L) concentrations in natural groundwater. The extent of sorption is influenced by colloid concentration, ionic strength, radionuclide concentration, and the presence of competing metals. Uranium (VI) and cesium sorption to bentonite colloids was investigated both separately and together in low ionic strength (2.20 mM) artificial rainwater (ARW) and high ionic strength (169 mM) artificial groundwater (AGW; representative of a fractured carbonate rock aquitard). Sorption experiments were conducted as a factor of colloid concentration, initial metal concentration and opposing metal presence. It was shown that both U(VI) and Cs sorption were significantly reduced in AGW in comparison to ARW. Additionally, the sorption coefficient  $K_d$  of both metals was found to decrease with increasing colloid concentration. Competitive sorption experiments indicated that at high colloid concentration (1–2 g/L), Cs sorption was reduced in the presence of U(VI), and at low colloid concentration (0.01–0.5 g/L), both Cs and U(VI)  $K_d$ s were reduced when they were present together due to competition for similar sorption sites. The results from this study imply that in brackish carbonate rock aquifers, typical of the Israeli northern Negev Desert, both U(VI) and Cs are more likely to be mobile as dissolved species rather than as colloid-associated solids.

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\* Corresponding author.

E-mail address: [weisbrod@bgu.ac.il](mailto:weisbrod@bgu.ac.il) (N. Weisbrod).

## 1. Introduction

Sorption of radionuclides to colloidal-sized clay particles has often been implicated in their increased transport through fractured rock aquifers (Missana et al., 2008; Schäfer et al., 2012; Tang and Weisbrod, 2010; Zhang et al., 2012). The extent and irreversibility of this sorption is dependent on many factors, including the pore water solution chemistry, pH and redox potential, ionic strength, and radionuclide concentration. Interactions between radionuclides and bentonite clay are of particular interest as bentonite is used in plans for deep geological repositories as both a backfill and buffer designed to impede the outward migration of leaked waste (Ali Khan, 2003; Bachmaf et al., 2008; Catalano and Brown, 2005; Galamboš et al., 2010; Geckeis et al., 2004; Missana et al., 2011; Wang et al., 2005; Zheng et al., 2017). Bentonite's high cation exchange capacity facilitates the sorption of radionuclides onto the backfill, thereby preventing their migration into surrounding aquifers. However, multiple studies have found that colloidal-sized particles eroded from the backfill can be transported readily through fracture systems in the subsurface and increase radionuclide mobility (Ali Khan, 2003; Missana et al., 2011; Norrfors et al., 2016; Zhang et al., 2012). Interactions with bentonite colloids, including both sorption and desorption processes, therefore play an important role in dictating the overall potential mobility of radionuclides in the subsurface.

Numerous studies on radionuclide sorption to bentonite colloids have been conducted at relatively high solid to solution ratios, ranging from several g/L to hundreds of g/L (e.g., Cheng et al., 2006; Deepthi Rani and Sasidhar, 2012; Miller et al., 2010; Missana et al., 2004). These studies use high bentonite concentrations to ensure that significant sorption of the radionuclides is observed, and thus any variations in sorption resulting from changing the pH, ionic strength or other factors can be accurately determined. However, deep groundwater natural colloid concentrations are generally quite low. At the Grimsel Test Site (Swiss Alps), for instance, deep groundwater colloid concentration was reported to be <1 mg/L (Degueldre et al., 1989). Another study reported low colloid concentration (0.12–0.029 mg/L) in drain water collected from an abandoned mine in Luxembourg (Filella et al., 2009). Previous work conducted in a field site close to where water from the present study was sampled has also revealed low particle concentrations (<10 mg/L) potentially capable of influencing the transport of sorbed contaminants (Weisbrod et al., 2002). High heterogeneity in particle concentration is sometimes observed even within a small area, such as within a single well throughout the water column of an aquifer (Weisbrod et al., 1996). It has been shown, albeit at a much greater solid mass range, that the sorption coefficient of U(VI) to goethite-coated sand was in fact highly dependent on the solid to solution ratio (Cheng et al., 2006). As colloid presence has been shown to influence reactive radionuclide transport and recovery at low colloid concentration of 20 mg/L (Möri et al., 2003), it is important to understand the effect of colloid concentration on radionuclide sorption properties to best predict potential radionuclide mobility.

An additional body of research focuses on radionuclide sorption and transport at low ionic strengths (Bachmaf et al., 2008; Catalano and Brown, 2005; Missana et al., 2004; Norrfors et al., 2016; Tran et al., 2016) at solid to solution ratios ranging from 0.02–20 g/L. While this is representative of several potential deep geological disposal sites, many aquifers, especially those in coastal regions (Re and Sacchi, 2017) and arid environments (Weisbrod et al., 2000), exhibit high salinities (Schmeide et al., 2016; Tanji, 2002), which have major implications for sorption and transport of heavy metals and radionuclides (Acosta et al., 2011; Rout et al., 2015) and consequently their potential mobility. Furthermore, high ionic strength groundwater has been reported in aquifers surrounding the WIPP (Waste Isolation Pilot Plant) in the USA, where deep subsurface salt beds are considered good candidates for nuclear disposal (Borkowski et al., 1996; Runde, 2000). Finally, high ionic strength groundwater may also result from soil salinization (Acosta et al., 2011) and sea water intrusion (Rout et al., 2015;

Werner et al., 2013). Therefore, two main types of water were chosen for investigation in this study. The first is an artificial rainwater (ARW) that contains the same ionic composition and strength (2.2 mM) as water used for colloid-facilitated transport experiments in previous studies (Tang and Weisbrod, 2010; Tran et al., 2015, 2016) and will allow for comparison of results from this study to previous work. Furthermore, the observation of sorption at comparatively low ionic strengths helps keep this work in the context of a larger body of literature presenting sorption in conditions of low ionic strength such as the glacial melt waters of the Grimsel Test Site (Schäfer et al., 2012). The second water investigated is an artificial groundwater (AGW) of high ionic strength (169 mM) that contains a similar ionic composition to water measured in a field site (supporting information, Table S1) sampled in the Northern Negev Desert, Israel. This second water type will allow for better prediction of the actual transport potential of leaked radionuclides in this region.

Cesium (Cs) and uranium (U) were chosen for investigation, as they may be found together in cases of radioactively contaminated sites, radioactive waste disposal and mine tailings (Seaman et al., 2001). Under oxidizing conditions, U is most stable in the (VI) oxidation state, which exists as the uranyl ion,  $\text{UO}_2^{2+}$  in solution. Uranyl's sorption to surfaces is highly influenced by its complexation with carbonate (Bachmaf et al., 2008; Kersting, 2012; Schmeide et al., 2016), which has been shown to impede sorption of  $\text{UO}_2^{2+}$  to montmorillonite (Bachmaf et al., 2008; Marques Fernandes et al., 2012). In the presence of calcium, it is known to form stable  $\text{Ca-UO}_2\text{-CO}_3$  ternary complexes in solution (Burow et al., 2017; Dong et al., 2005; Dong and Brooks, 2006; Vercouter et al., 2015). In the majority of cases, uranyl sorption to mineral surfaces is dominated by surface complexation (Marques Fernandes et al., 2012; Payne et al., 2004; Tournassat et al., 2018; Zheng et al., 2017). However, it has also been reported that at low ionic strengths and circumneutral pH, uranyl ions are, in fact, likely to sorb through cation exchange to edge sites on montmorillonite (Catalano and Brown, 2005). This may put it in direct competition with other radionuclides which sorb through a similar mechanism.

In contrast to the uranyl ion  $\text{UO}_2^{2+}$ , cesium ( $\text{Cs}^+$ ) ions do not react readily to form complexes in solution, and instead it has been shown to sorb quickly onto inorganic surfaces (Deepthi Rani and Sasidhar, 2012; Fuller et al., 2014; Missana et al., 2004; Seaman et al., 2001; Tang and Weisbrod, 2010). Though Cs sorption to clays is typically thought of as occurring nearly exclusively through cation exchange (Cherif et al., 2017), Wang et al. (2005) also showed that at high pH,  $\text{Cs}^+$  may sorb to bentonite by surface complexation. This means that in certain instances, Cs and U(VI) may be in direct competition for similar sorption sites. The impact of competing radionuclide presence on sorption  $K_d$  has rarely been investigated, even though radionuclides are unlikely to be found as individual elements and may influence each other's sorption properties (Gutierrez and Fuentes, 1993). Therefore, in addition to investigating radionuclide sorption in binary systems (a single mineral and single radionuclide), it is also important to understand sorption characteristics in ternary systems, where more than one radionuclide is present.

In this study, the sorption of Cs and U(VI) to bentonite clay colloids under carbonate rich conditions has been investigated as a function of ionic strength (expressed as matrix water type), colloid concentration and initial metal concentration. Additionally, the sorption of the two radionuclides in a ternary system as a factor of colloid concentration was explored. These experiments allow for a prediction of the maximum potential sorption of radionuclides to bentonite colloids under the conditions explored, and are designed to demonstrate the impact of salinity, carbonate and competing radionuclide presence on sorption processes. Experiments were designed using salt concentrations mimicking those measured in the northern Negev Desert, Israel, such that results can be used to predict U(VI) and Cs transport under site-specific geochemical conditions.

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