

Linearity and reversibility of iodide adsorption on sediments from Hanford, Washington under water saturated conditions[☆]

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Abstract

A series of adsorption and desorption experiments were completed to determine the linearity and reversibility of iodide adsorption onto sediment at the Hanford Site in southeastern Washington. Adsorption experiments conducted with Hanford formation sediment and groundwater spiked with dissolved ¹²⁵I (as an analog tracer for ¹²⁹I) indicated that iodide adsorption was very low (0.2 mL/g) at pH 7.5 and could be represented by a linear isotherm up to a total concentration of 100 mg/L dissolved iodide. The results of desorption experiments revealed that up to 60% of adsorbed iodide was readily desorbed after 14 days by iodide-free groundwater. Because iodide adsorption was considered to be partially reversible, even though small amount of initial iodide is retarded by adsorption at mineral–water interfaces, the weak adsorption affinity results in release of iodide when iodide-free pore waters and uncontaminated groundwaters contact the contaminated sediments in the vadose zone and aquifer systems.

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1. Introduction

Nuclear wastes generated from Pu production and separation processes of nuclear materials are currently being stored in underground storage tanks at the US Department of Energy's (DOE) Hanford Site in southeastern Washington state [1,2]. Current plans in the DOE immobilized low-activity waste (ILAW) program are for the low-activity radioactive wastes to be vitrified and then stored in near surface burial facilities at the 200 East Area of the Hanford Site [1]. Previous performance assessment (PA) analyses of the proposed ILAW disposal site have shown that ¹²⁹I, ²³⁷Np, ⁷⁹Se, ⁹⁹Tc,

and ^{233,235,238}U pose the most potential risk to human health [1]. Because the severity of environmental problems associated with these radionuclides depends on how readily these contaminants migrate from the source through the accessible environments, the risks associated with relatively mobile radionuclides are a great concern [3].

Radioactive iodine has been released into soils and the atmosphere during the course of nuclear power plant operations, nuclear weapon tests, nuclear accidents such as that at the Chernobyl power plant, and operations and disposal activities associated with nuclear-fuel reprocessing [4–8]. Health risks associated with the release of radioactive iodine, especially as ¹²⁹I, are a significant environmental concern because of the long-half life of ¹²⁹I ($t_{1/2} = 1.6 \times 10^7$ years) and high mobility of all iodine isotopes in subsurface environments.

Previous studies have reviewed the environmental behavior of iodine [3,9–11]. Iodine is known to occur in

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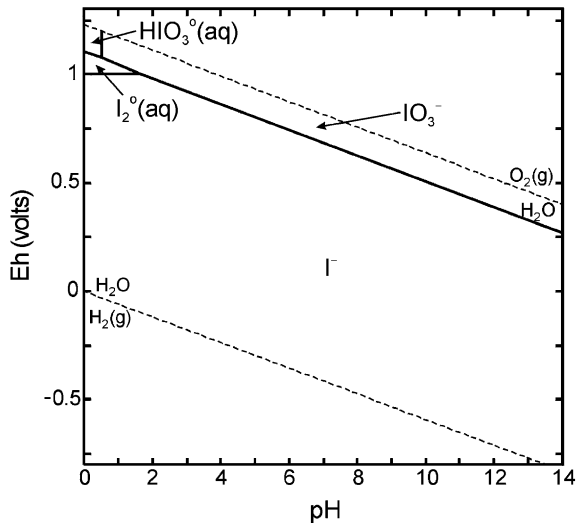


Fig. 1. Eh-pH stability diagram for dominant iodine aqueous species at 25°C. Diagram was calculated using the Geochemist's Workbench[®] 4.0 software package based on a total concentration of 10^{-8} mol/L dissolved iodine. The dashed lines represent the boundaries for the thermodynamic stability of water.

several oxidation states (Fig. 1). In most aqueous environments, iodine is present in the -1 valence state as the iodide ion, I^- . The stability range of I^- extends almost over the entire pH and Eh range for the thermodynamic stability of water. In marine and highly oxidizing environments such as surface waters and some highly oxygenated shallow groundwaters, iodine may be present in the $+5$ oxidation state as the iodate ion, IO_3^- . Under highly oxidizing conditions at pH values less than 4, molecular I_2° (aq) may form from the reduction of IO_3^- or oxidation of I^- .

Due to the inherent negative charge of most mineral surfaces at near neutral and basic pH conditions, dissolved anions, such as I^- , are electrostatically repelled by sediment minerals and show relatively weak adsorption behavior, suggesting mobile transport in sediment systems at these pH conditions. Geochemical literature indicates that iodide adsorption is affected by the organic matter content, iron-oxide concentration, and pH conditions of sediments [12–14]. In general, iodide adsorption on bentonite and hematite [13], calcite [14], and montmorillonite [15] was reported to be negligible at neutral and basic pH conditions. While a high distribution coefficient (K_d) value for iodide was found onto illite [16], most of the reported K_d values for iodide on other adsorbents including clay minerals are typically low at basic pH conditions.

Because iodide K_d values are sensitive to geochemical parameters such as pH, soil mineralogy, and organic carbon content, site-specific adsorption/desorption experiments for iodide are required to measure credible K_d

values used to predict the fate and transport of radioactive iodide for each specific site risk assessment. The K_d construct assumes that adsorption is an equilibrium process, independent of adsorbate concentration (i.e., linear adsorption isotherm), and totally reversible [17]. However, K_d values will deviate from linearity when the adsorption capacity of sediment is approached [17]. Moreover, some studies have shown that the adsorbed solutes may not be readily desorbed or released from the mineral surfaces back into the aqueous phase [18]. The main objectives of this study were to experimentally determine the linearity of iodide adsorption as a function of iodide concentration and the extent of adsorption reversibility of iodide for geochemical conditions similar to the Hanford site.

2. Materials and methods

2.1. Materials

The composite sediment used in this study as the adsorbent was collected from the ILAW borehole #2 (well name 299-E24-21; borehole number C3177) at the 200 East Area of the Hanford Site. Particle size distribution was determined by the wet sieve/hydrometer method [19]. Based on the modified Folk–Wentworth [20,21] classification, the composite sample consisted of 3.9 wt% gravel, 88.6 wt% sand, 6.2 wt% silt, and 1.4 wt% clay. The X-ray diffraction (XRD) analyses showed that quartz and feldspar were dominant minerals with lesser amounts of mica, chlorite, and an amphibole, which is consistent with other reported XRD analyses of Hanford Site sediments [16]. Smectite, chlorite, illite, and kaolinite were also identified in the clay-size fraction using XRD.

The sediment pH was measured using a 1:1 ratio of sediment to distilled water. A mean sediment pH of 7.5 was calculated from the results of duplicate measurements. A surface area of $41.8 \text{ m}^2/\text{g}$ was measured for the clay-size fraction ($<1.4 \mu\text{m}$) using a Micromeritics ASAP 2010 surface analyzer and the BET- N_2 method [22].

The composite sediment used in the adsorption–desorption experiments was sieved to remove gravel particles greater than 2 mm. Uncontaminated groundwater collected from well 699-S3-25 at the Hanford Site was used as a background solution in all experiments. Table 1 lists the composition of the groundwater as analyzed using standard techniques. All experiments were conducted at ambient temperature (20–22°C) using NANOpure™ water (reagent-grade water with at least $18 \text{ M}\Omega\text{cm}$ resistivity) and reagent-grade chemicals. Because the experimental protocol was not designed to remove the effects of dissolved $\text{CO}_2(\text{g})$, all experiments were conducted in test tubes closed to the atmosphere

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