



Sorbent materials for rapid remediation of wash water during radiological event relief



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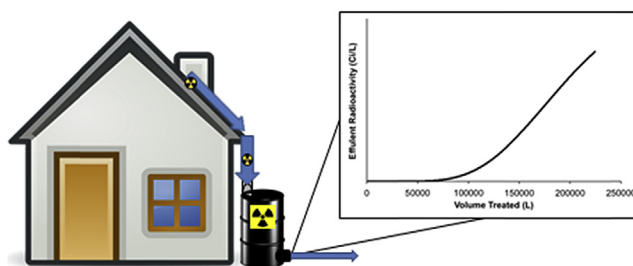
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HIGHLIGHTS

- Vermiculite and montmorillonite are proposed for use in retention barrels.
- Retention barrels retain radionuclides while allowing for wash water flow.
- Vermiculite demonstrated a high selectivity for ¹³⁷Cs.
- Montmorillonite displayed the ability to sorb low concentrations of ⁸⁵Sr.

GRAPHICAL ABSTRACT



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ABSTRACT

Procedures for removing harmful radiation from interior and exterior surfaces of homes and businesses after a nuclear or radiological disaster may generate large volumes of radiologically contaminated waste water. Rather than releasing this waste water to potentially contaminate surrounding areas, it is preferable to treat it onsite. Retention barrels are a viable option because of their simplicity in preparation and availability of possible sorbent materials. This study investigated the use of aluminosilicate clay minerals as sorbent materials to retain ¹³⁷Cs, ⁸⁵Sr, and ¹⁵²Eu. Vermiculite strongly retained ¹³⁷Cs, though other radionuclides displayed diminished affinity for the surface. Montmorillonite exhibited increased affinity to sorb ⁸⁵Sr and ¹⁵²Eu in the presence of higher concentrations of ¹³⁷Cs. To simulate flow within retention barrels, vermiculite was mixed with sand and used in small-scale column experiments. The GoldSim contaminate fate module was used to model breakthrough and assess the feasibility of using clay minerals as sorbent materials in retention barrels. The modeled radionuclide breakthrough profiles suggest that vermiculite-sand and montmorillonite-sand filled barrels could be used for treatment of contaminated water generated from field operations.

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1. Introduction

Atmospherically-deposited radionuclides, resulting from a nuclear or radiological disaster (i.e. nuclear reactor meltdown, dirty

bomb), should be rapidly removed from interior and exterior surfaces of homes and businesses to reduce exposure (Sinkko et al., 2004; Ring, 2004; Conklin and Edwards, 2000; Levenson and Rahn, 1981; Paton and Johnston, 2001; Kaminski et al., 2016a). Radionuclides can be disseminated long distances in contamination plumes from the disaster site (Evrard et al., 2013; Simmonds et al., 1982). Of the long lived radionuclides, ¹³⁷Cs is generally the most abundant, making it a primary concern in mitigation procedures

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(Simmonds et al., 1982; Andersson, 2009; Konoplev and Bobovnikova, 1991; Bunzl et al., 1992). In addition, ^{90}Sr and low levels of transuranic radionuclides (i.e. ^{241}Am) have been detected in nuclear fallout and should be considered when developing practices (Bunzl et al., 1992; Shinonaga et al., 2014). Initially, the removal of dust and debris is prescribed as an early course of action to remove radioactive particulate matter from surfaces (Dick and Baker, 1961; Nisbet et al., 2009, 2010). However, wet deposited radionuclides, especially those that are positively charged (^{137}Cs , ^{90}Sr , ^{241}Am), are subject to ion exchange reactions with building materials (Andersson, 2009; Samuleev et al., 2013; Thiessen et al., 2009). Exchanged radionuclides can be removed by washing building surfaces with water or concentrated salt solutions (Andersson, 2009; Samuleev et al., 2013; Thiessen et al., 2009). Washing procedures can result in a large quantity of contaminated wastewater, which, if left untreated, can spread radiological contamination to surrounding areas (Sinkko et al., 2004; Kaminski et al., 2016a, 2016b). A potential technique to manage this wash water is the treatment of dissolved radionuclides using sorbent materials located inside retention barrels. These barrels would contain sorbent materials that retain dissolved radionuclides while allowing wash water to flow through. The barrels with contaminated solids can then be efficiently transported due to their compact nature and properly disposed. The sorbents utilized within the barrels must be widely available, economical, and have a high selectivity to sorb dissolved radionuclides from backgrounds containing other inorganic cations.

Aluminosilicate clay minerals are naturally abundant, widely available, minerals that often have high, non-pH dependent, cation exchange capacity (CEC) and a high selectivity for cationic radionuclides (Staunton and Roubaud, 1997; Matocha, 2006). For example, illite has high specificity to sorb ^{137}Cs from aqueous solutions due to a collapsed interlayer and a higher amount of isomorphous substitutions in the silicon oxide layer (Comans and Hockley, 1992; Comans et al., 1991). The silicon oxide layer substitutions are located closer to the surface for 2:1 clays; which decreases the area over which the charge is spread, focusing the charge over only three adjacent surface oxygen atoms (Matocha, 2006; Levy and Shainberg, 1972; Ras et al., 2007). The interaction energy is far greater between focused charge sites and ions that can shed hydration shells, creating a natural selectivity for ^{137}Cs or similar ions (Matocha, 2006; Levy and Shainberg, 1972; Ras et al., 2007). However, this process occurs within the collapsed interlayer, making it kinetically slow compared to electrostatic interactions between dissolved cationic radionuclides and other clays (Comans and Hockley, 1992; Comans et al., 1991; Poinssot et al., 1999). Therefore clay minerals without a collapsed interlayer may be preferable as sorbents deployed in retention barrels, since contact time is limited in flow through applications.

The fully hydrated interlayers of vermiculite and montmorillonite clay minerals may result in fewer limitations due to kinetic effects during cationic radionuclide uptake (Wu et al., 2009; Hadadi et al., 2009). The main differences between vermiculite and montmorillonite are the abundance and placement of isomorphous substitutions (Matocha, 2006; Levy and Shainberg, 1972; Ras et al., 2007). Montmorillonite has more aluminum oxide substitutions, resulting in less focused charge sites and CECs ranging from 75 to 90 meq/100 g (Matocha, 2006; Levy and Shainberg, 1972; Ras et al., 2007). Sorption coefficients to montmorillonite can vary drastically for cationic radionuclides depending on the identity and abundance of background cations (Staunton and Roubaud, 1997; He and Walling, 1996; Atun et al., 1996). For instance, sorption coefficients of ^{137}Cs onto montmorillonite with calcium as the dominant ion are 10 times lower than those with potassium or sodium as the dominant exchange ion at background concentrations lower than

10 mM (Staunton and Roubaud, 1997). Vermiculite, on the other hand, displays less of these effects as most of the substitutions occur in the silicon oxide layer near the surface similar to illite (Matocha, 2006; Levy and Shainberg, 1972; Ras et al., 2007; Sawhney, 1972; Tamura, 1961). The high energy of interaction between focused charge groups of vermiculite and ^{137}Cs increases the selectivity of the clay (Sawhney, 1972). However this selectivity decreases the sorption coefficient for other radionuclides on vermiculite, including ^{90}Sr and ^{241}Am , especially in the presence of ^{137}Cs (Konishi et al., 1988; Sljivic-Ivanovic et al., 2015). Lower sorption coefficients indicate decreased retention of these radionuclides if vermiculite is the clay mineral deployed in the retention barrels. Additionally, the swelling nature of montmorillonite and vermiculite decreases their hydraulic conductivity, restricting the flow in the retention barrel (Rowe et al., 2004). A possible, readily available, solution to this problem is mixing the clay minerals with sand (which has high hydraulic conductivity but low CEC) to allow both flow and radionuclide retention. The presence of such a mixture, however, greatly complicates retention barrel design. Predictive models must be used to initially assess operational parameters, such as the relative amount of aluminosilicate minerals and sand needed in a retention barrel.

This study aims to evaluate vermiculite and montmorillonite as possible sorbent materials for use in retention barrels prior to large-scale testing. Removal of ^{137}Cs was the primary goal, however, ^{85}Sr , and ^{152}Eu were also studied. ^{85}Sr and ^{152}Eu were served as radioactive surrogates for ^{90}Sr and ^{241}Am , respectively, because of detection and waste classification issues. ^{152}Eu is documented as an adequate surrogate for ^{241}Am in regards to sorption to geologic minerals (Lee et al., 2006). Radiochemicals were exclusively utilized to enable low contaminant concentrations without detection limit issues and to fully account for competition effects with other salts present in retention barrel water. The Goldsim contaminant fate module was employed to predict field-scale operational parameters.

2. Experimental

2.1. Materials

^{137}Cs , ^{85}Sr , and ^{152}Eu were obtained from Perkin Elmer (MA, USA). Vermiculite was from Specialty Vermiculite (Grace vermiculite: SC, USA) and the Strong Company (Strong vermiculite: AR, USA). Montmorillonite was from the American Colloid Company (Volclay 205x: IL, USA), Acros Organics (K10 montmorillonite: MA, USA) and the Clay Minerals Society (Wyoming montmorillonite: Wyoming, USA). Sand was obtained from New Plant Life (IA, USA). All solids were used as received. Tap water (pH 7.5 ± 0.3) is supplied and routinely analyzed at the DuPage County Department of Public Works. At the time of the experiments a sample was analyzed for metals by ICP-MS (Table 1).

2.2. Batch experimental methods

Sorption experiments to all clays were performed for ^{137}Cs with a solid-to-water ratio of 1 mg/mL. 1.5 mL reactors were set up in duplicate with initial ^{137}Cs radioactivities of 824 $\mu\text{Ci/L}$ (0.07 μM). Initial ^{137}Cs concentrations were designed to keep sorbed concentrations well below 1% surface coverage of the clay with the lowest CEC (Wyoming montmorillonite: 0.76 meq/100 g) while keeping aqueous concentrations above the limit of detection (30 $\mu\text{Ci/L}$). Sorbed concentrations below 1% of the CEC have previously shown not to be influenced by isotherm non-linearity, thus keeping point sorption coefficients within the linear range (Adeleye et al., 1994; Cornell, 1993). Further, wash waters generated from mitigation

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