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Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

Collection of fission and activation product elements from fresh and ocean waters: A comparison of traditional and novel sorbents

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ARTICLE INFO

Article history:

Received 19 February 2010

Received in revised form

16 July 2010

Accepted 30 July 2010

Keywords:

Chemisorption

Sorbents

Resins

Nanomaterials

Radionuclides

Fission products

ABSTRACT

Monitoring natural waters for the inadvertent release of radioactive fission products produced as a result of nuclear power generation downstream from these facilities is essential for maintaining water quality. To this end, we evaluated sorbents for simultaneous in-situ large volume extraction of radionuclides with both soft (e.g., Ag) and hard metal (e.g., Co, Zr, Nb, Ba, and Cs) or anionic (e.g., Ru, Te, Sb) character. In this study, we evaluated a number of conventional and novel nanoporous sorbents in both fresh and salt waters. In most cases, the nanoporous sorbents demonstrated enhanced retention of analytes. Salinity had significant effects upon sorbent performance and was most significant for hard cations, specifically Cs and Ba. The presence of natural organic matter had little effect on the ability of chemisorbents to extract target elements.

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1. Introduction

Monitoring water quality in proximity of historic and modern nuclear facilities is essential for maintaining environmental and human health (Egorov et al., 2006; Grate et al., 2008). Fission products produced during nuclear power generation include hundreds of different isotopes with half-lives ranging from billions of years to fractions of seconds (Shapiro, 1993). In addition, legacy wastes, such as the 53 million gallons contained in the 177 underground storage tanks at the Hanford Nuclear Reservation (Adu-Wusu et al., 2006), require long-term stewardship. Accidental releases of these wastes to marine, coastal, and fresh waters have been documented in the past (Santschi et al., 1990) and risk of future releases may increase with the advent of the global nuclear renaissance. Continuous monitoring of water bodies near these sites could provide early detection of radioactive contaminant releases as they occur.

To help meet current and future environmental monitoring needs around nuclear facilities and waste sites, a medium-resolution, autonomous, in-situ gamma detection system is being developed for monitoring marine and coastal waters for gamma-emitting fission and activation products (Schwantes et al., 2009).

The primary novelty of this system is that gamma-emitting isotopes of interest are preferentially extracted from natural waters prior to their detection in order to minimize background from naturally occurring radioactive isotopes (including, ⁴⁰K and U/Th series radionuclides) and eliminate signal attenuation of the gamma rays traveling through water (Ho et al., 2005). It is estimated that preconcentrating prior to detection will afford one to three orders of magnitude lower limits of detection over conventional direct detection (Schwantes et al., 2009). Filtration is used to preconcentrate target isotopes residing on suspended particles, while chemisorption is employed to preferentially extract more dissolved components from the water column. For preconcentrating dissolved components of interest, a variety of conventional and proprietary nanoporous chemisorbents are being considered for use.

Characteristics of good chemisorbents include high analyte affinity, good chemical selectivity a large capacity for target elements of interest, stability of performance over a wide range of pH and ionic strengths, and adequate permeability. The latter characteristic minimizes issues related to clogging when passing large volumes of water across the preconcentrating sorbent. Highly selective sorbents are important since natural waters contain a number of confounding constituents (including proteins, organic substances, surfactants, cations, and anions) that can compete with the target analytes for binding sites on the chemisorbent (Lin et al., 2005; Yantasee et al., 2007, 2008).

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Table 1
Typical environmental concentrations, speciation, and distribution properties of target analytes in fresh and seawater.

Element	Stable oceanic conc. (M) ^a	Stable river conc. (M) ^a	Typical species and oxidation state in natural waters ^a	Species and oxidation state of tracer	Typical log K_D (L/kg)	Metal type ^b
Co	2.0E–11	3.5E–09	II (CoCO ₃) ⁰	II (CoCl ₂)	5 ^a	A, B
Zr	3.0E–11	–	IV (Zr(OH) ₅ [–])	II (Zr(NO ₃) ₂)	4–5 ^c	A
Ru	5.0E–14	4.7E–09	VI (RuO ₄ ^{2–}) II (RuNO ₃ ⁺)	III (RuCl ₃)	3 ^c	A, C
Ag	3.5E–11	9.3E–12	I (AgCl ₂ [–])	I (AgNO ₃)	5 ^d	B
Te	5.0E–12	2.4E–11	I (AgCl ₃ ^{2–}) ^e VI (TeO(OH) ₅ [–]) VI (Te(OH) ₆ ⁰) IV (TeO(OH) ₃ [–])	VI (TeCl ₆)	~3	C
Sb	1.2E–09	5.8E–10	V (Sb(OH) ₆ [–]) III (Sb(OH) ₃ ⁰) ^e	III (SbCl ₃)	2 ^f	C
Ba	1.0E–07	4.4E–07	II (Ba ²⁺)	II (BaCl ₂)	2.5 ^f	A
Cs	2.2E–09	2.6E–10	I (Cs ⁺)	I (CsCl)	2.8 ^f	A

^a Elemental values, taken from Fig. 3.1, page 98–99, Millero (1996) and Li (2000).

^b Type A (hard) and type B (soft) metal ions, according to Pearson (1963) and type C=primary anionic species according to Nieboer and Richardson (1980).

^c Duursma and Gross (1971).

^d Wen et al. (1997).

^e Byrne (2002).

^f Li et al. (1984).

Sorbents of high selectivity maintain the ability to preconcentrate target species even in the presence of excess interfering cations and/or competing ligands (Lin et al., 2005).

Numerous conventional resins, ion exchangers and mineral precipitates (e.g., manganese dioxide, iron oxide, CuFeCN), and other sorbent materials have been widely applied for sequestering metals from aqueous solutions (Baskaran et al., 1993, 2009 and references therein). Procedures outlining the production of a number of these materials are available from the literature (Prout et al., 1965; Watari et al., 1967), while others are available commercially (Bandong et al., 2001; Peterson et al., 2006). In addition to these conventional chemisorbents, researchers at Pacific Northwest National Laboratory have developed novel nanoporous functionalized chemisorbents. These self-assembled monolayers on mesoporous supports (SAMMS^{TM1}), functionalized with similar reactive surfaces as the conventional resins, can afford exceptional high sorption capacities without significant loss in flow characteristics during large volume sampling (Fryxell et al., 2007). Some of the surface chemistries have been shown to perform well under a wide range of environmental conditions, particularly in waters with high ionic strength. For this reason, a study was undertaken to assess the performance of conventional and novel SAMMS chemisorbents for sequestering select fission and activation product elements in synthetic and real fresh and saline waters.

The SAMMS materials have a rigid open pore structure with an extremely high surface area (up to 480 m²/g) (Lin et al., 2001). Over 99.9% of the surface area of SAMMS available to solution borne reagents resides inside the honeycomb structure. Previous studies have found kinetics during sorption to these surfaces to be reaction limited and not restricted by diffusion into and out of the pores (Fryxell et al., 2005, 2007; Lin et al., 2001). SAMMS monolayers can be tailored to capture heavy metals, actinides, lanthanides, oxometallate anions, cesium, and iodine (Yantasee et al., 2008). Thiol functionalized SAMMS (SH-SAMMSTM) have demonstrated large binding site density 2.8 mmol SH g^{–1}, and partition coefficients greater than 10⁶ (mL/g) for Cu, Ag, Hg, Cd, Pb, in a variety of waters (Feng et al., 1997; Yantasee et al., 2007, 2008). Actinide

ligand functional groups, such as hydroxypridinone (HOPO) bind strongly with large actinide cations when functionalized onto SAMMS (Fryxell et al., 2005). Copper ferrocyanide functionalized SAMMS, including Cu-ferrocyanide-ethylenediamine-SAMMS, have demonstrated rapid kinetics and high capacity for Cs, with 99% of the uptake completed within 1 min and an estimated loading capacity of 1.35 mmol Cs per g of sorbent (Lin et al., 2001).

In this study, we competitively evaluated a number of sorbent materials for the collection of a diverse range of fission and activation product elements of the nuclear fuel cycle including Co, Zr, Nb, Ru, Ag, Te, Sb, Ba, and Cs. Selection of metals for testing of the chemisorbents was also based on representative species of A and B type metals, and C-type metalloid anionic species (see Table 1). Type A metals strongly bind with oxides, carbonates, and, at times, phosphates, while type B metals have been found to have a high affinity for sulfur groups. Transition metals have characteristics in between that of the type A and B metals (Pearson, 1963, and others). Capture performance of novel nanostructured sorbents was compared to conventional chemisorbents in both fresh, riverine, and saline, oceanic, waters.

2. Materials and experimental methods

2.1. Waters

Two synthetic and two natural waters were used for testing the performance of chemisorbents. The two synthetic waters (no organics) were produced and used to represent fresh (salinity=0.06%) river water and seawater (salinity=35%). Characteristics of each are given in Table 2. Artificial waters were buffered during the kinetic experiments using 50 mM Tris/25 mM HCl, which was later found to increase the salinity by approximately 3%. The pH of the buffered solutions was maintained within ± 0.1 pH units of 8.1 over the duration of the experiments. Without buffering, the pH of the artificial sea and river waters were approximately 7.8 and 9.1. Addition of buffering salts and percolating the synthetic freshwater solutions with high-purity CO₂ gas reduced the pH of each solution to approximately 8.0.

Natural waters included fresh river waters taken from the Columbia River, collected near Richland, Washington, and coastal

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