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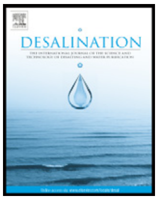

Removal of low level americium-241 from potable water originated from different geochemical environments by calcium alginate

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ABSTRACT

In a nuclear accident eventuality, the decontamination of potable water without disturbing the water quality is one of the major challenges. During this work, efforts have been made to reduce the contamination of americium (Am)-241 from potable water effectively by using calcium alginate followed by treatment with activated charcoal. Laboratory simulated experiments were carried out for the removal of low level of Am-241 from the potable water originated from groundwater, rain water and lake water. Water samples after filtering through 0.45 μm filter paper were spiked in the range of 10–200 Bq L^{-1} of Am-241 as $Am(NO_3)_3$. In the pH range 1–8, 60–98% Am was sorbed in the beads with a maximum observed at pH 4 ± 0.2. It was observed that the variation of carbonate, bicarbonate, does not influence the sorption capacity of calcium alginate whereas the DOC decreased the sorption capacity in the range of 40–93% depending on DOC concentration. Ninety-eight to ninety-nine percent of Am is recovered from sodium alginate beads by using 0.6 M HNO_3. The mechanism of interaction of Am with calcium alginate is proposed based on attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR).

1. Introduction

In 2008, WHO published a third edition of the Guidelines for Drinking Water Quality. These guidelines are not intended to provide a control on amounts of activity which may be discharged into drinking water sources, but are guidelines from which drinking water suppliers can determine whether action is necessary to reduce radioactivity levels in the water supply (WHO, 2008). These levels relate to natural and artificially enhanced radioactivity in drinking water. The guidelines present a two-stage approach. Screening levels of 0.1 Bq L^{-1} and 1 Bq L^{-1} for gross alpha and gross beta activities respectively have been retained from the first edition. If activity of drinking water is below this level, no further action is required [1]. Allowable concentration of Am-241 in drinking water as per guidelines of WHO issued in 2008 is 11 Bq L^{-1} [2].

Radioactive constituents of drinking-water can result from the following:

- Naturally occurring radioactive species (e.g., radionuclides of the thorium and uranium decay series in drinking-water sources), in particular radium-226/228 and a few others;
- Technological processes involving naturally occurring radioactive materials (e.g., the mining and processing of mineral sands or phosphate fertilizer production).
- Radionuclides discharged from nuclear fuel cycle facilities.
- Manufactured radionuclides (produced and used in unsealed form), which might enter drinking-water supplies as a result of regular discharges and, in particular, in case of improper medical or industrial use and disposal of radioactive materials.
- Past releases of radionuclides into the environment, including water sources.

Alginate is the most abundant marine biopolymer. The major source of alginate is found in the cell walls and the intracellular spaces of brown seaweed. Alginate is a sodium salt of alginic acid and provide the plant with both flexibility and strength, which are necessary for plant growth in the sea. Alginic acid is an unbranched binary copolymer of 1–4 glycosidically linked -L-guluronic acid (G) and its C-5 epimer -D-mannuronic acid (M) [3–5]. The salts (and esters) of this polysaccharides are generally named alginates. The sodium alginate is generally used in various food items like jelly and hence its negative impact on the water quality is minimal. The use of alginate as an immobilizing agent in most applications rests in its ability to form heat-stable strong gels which can develop and set at room temperatures. It is the alginate gel formation with calcium ions which has been of interest in most applications.

A large number of studies has already been conducted to exhibit excellent adsorption ability for multivalent metal ions by calcium alginate beads [6–8]. The ability of alginate to form gels by ion-exchange reaction with multivalent metal ions suggests its use as a metal adsorbent. Many recent studies have addressed the collection of

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heavy metals such as Co, Cu, Cd and Zn by alginic acid [9–12]. The alginic acid and alginates are thus likely to be potentially useful as adsorbents for the removal of radioactive nuclides from waste solutions. However, a limited amount of information is available concerning the uptake behavior of radioactive nuclides on these polymer gels from potable water. In the present study, decontamination of potable water from Am-241 without disturbing the water qualities has been done using sodium alginate beads followed by the treatment with activated charcoal column. Major water quality parameters were monitored to ensure the water quality.

2. Materials and method

2.1. Sample collection

The water samples were collected from different locations spread over a distance of 100 km^2 around Trombay, Mumbai having latitude 19°00′498 (N) and longitude 72°55′136. The samples were collected in 5 L polypropylene bottles. Various physicochemical characteristics like pH, conductivity, dissolved organic carbon (DOC), major and trace elements etc. were studied. The radioactive content was determined by measuring the gross alpha and beta radioactivities.

2.2. Preparation of simulated contaminated ground water solution

The water samples from three different origins (well water, rain water and lake water) were filtered through 0.45 μm filter paper by using suction filtration. The concentration of carbonate and bicarbonate was increased by using 2 M solution of $MgCO_3$ and $NaHCO_3$. Different water samples were spiked with the standard stock solution of Am as $Am(NO_3)_3$. Contaminated water was kept for a week under N_2 atmosphere for equilibration.

2.3. Formation of calcium alginate beads

A 2% solution (wt./vol.) of Na alginate in water was prepared with deionized water. This solution was taken in syringe and added to 0.2 M $CaCl_2$ solution drop-wise. Na alginate reacted with $CaCl_2$ to form beads of Ca alginate. These beads were stored in 0.2 M $CaCl_2$ solution and were thoroughly washed with deionized water before use. Calcium alginates beads so formed are stable in the temperature range 20–50 °C. The beads were permeable to water and had enough mechanical strength to be held in water column for 3–4 weeks.

2.4. Calcium alginate beads and granular activated charcoal column

Calcium alginate and granulated activated charcoal column was prepared by putting 20 g (wet weight) of calcium alginate beads and 10 g of granulated activated charcoal respectively in 20 cm long × 0.8 cm diameter glass columns separately. In case of calcium alginate, experiments were carried out in locally designed columns having a cup [diameter 36 mm which can hold a volume of 1 L] on top. Before elution from the calcium alginate column holdup time of 200 minutes was given. The eluted solutions from calcium alginate columns were passed through activated charcoal column. The rate of elution was maintained at a speed of 2 mL $minute^{-1}$ in both of the columns.

2.5. Measurement of physicochemical characteristics and concentration of major and trace metal ions

Various physiochemical characteristics like pH, conductivity, and redox potential were measured by using pH meter (Model $PICO^+$, Lab India) and conductivity meter (Model $PICO^+$, Lab India). In case of redox potential measurement single pin platinum electrode was used instead of single pin glass electrode for pH measurement. The concentration of various metal ions were measured by either Atomic Absorption Spectrometry (AAS) or Inductive Coupled Plasma-Optical Emission Spectroscopy (ICP-OES). The calibration for all the elements was carried out by using Fluka standards in HNO_3 medium.

2.6. Measurement of Am-241

The measurement of alpha activity of Am-241 was carried out by using ZnS(Ag) detector having a background of 0.07 cpm and counting efficiency of 39%. The efficiency of the system was evaluated by using Am-241 having an activity of 83.33 Bq. This electrodeposited source was prepared by using standard solution of Am-241 received from IAEA, discussed in detail elsewhere [1]. This electrodeposited source was calibrated against mixed nuclide source Am-241 from Amersham (AMR 33).

2.7. Measurement of Eu^{3+}

The determination of europium was carried out using an Inductively Coupled Plasma Optical Emission Spectrometer having a resolution of 19 pm with a grating of 2400 grooves per mm (ICP-OES, model JY 2000). The intensity of emission was measured at two different wavelengths i.e. 381.965 and 318.967 nm. The calibration of instrument was done between 1 and 10 μg L^{-1} by using Aldrich standard solution of Europium having strength of 1 g L^{-1}.

2.8. Attenuated total reflectance-Fourier transform infrared spectroscopy (ATR-FTIR)

ATR-FTIR spectrum of calcium alginate beads was recorded at various stages by using Fourier transform spectrometer “Spectrum One (Bruker)” equipped with the universal ATR as an internal reflection accessory having composite Zinc Selenide (ZnSe) and Diamond crystal. The Spectra was recorded in the wavelength range of 500–4000 cm^{-1} with a spectrum resolution of 4 cm^{-1}.

3. Results and discussion

The important water quality parameters monitored of the potable water collected from three different sources are given in the Table 1. From this table it is clear that except total hardness which is the measure of concentration of calcium and magnesium carbonate and total dissolved solid (TDS), there is not much variation in concentration of different parameters. There is a slight variation in the concentration of nitrate but still much below than the permissible limit of 10 μg mL^{-1} [2].

Table 1
Characteristics of potable water collected from different sources.

Parameters	Average value (μg L^{-1})		
	Ground water	Rain water	Lake water
pH	7.8 ± 0.4	4.5 ± 0.3	5.0 ± 0.4
Conductance (μS)	400 ± 30	200 ± 20	240 ± 21
Total hardness	100 ± 10	70 ± 8.7	72 ± 6.2
HCO_3^-	70 ± 6	30 ± 4	34 ± 2.3
Dissolved organic carbon (DOC)	1.5 ± 0.02	0.8 ± 0.08	2.30 ± 0.12
Total dissolved solid	180 ± 20	80 ± 10	100 ± 5
Turbidity (NTU)	7 ± 2	3 ± 1	5 ± 1
PO_4^{3-}	0.45 ± 0.03	0.25 ± 0.08	0.25 ± 0.06
NO_3^-	6 ± 0.8	2.2 ± 0.8	3.8 ± 0.8
F^{-1}	0.09 ± 0.01	0.05 ± 0.01	0.03 ± 0.01
Calcium	39.26 ± 3.20	27 ± 1.7	25 ± 1.4
Magnesium	17.2 ± 1.2	15 ± 1.1	15 ± 1.3
Arsenic (ng L^{-1})	<5	<5	<5
Cadmium	<0.01	<0.01	<0.01
Gross alpha (Bq L^{-1})	<0.1	<0.1	<0.1
Gross beta (Bq L^{-1})	<1.1	<1.1	<1.1