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Development of Natural Sorbent for Decontamination of Drinking Water from Low Level of Transuranic and Other Long-lived Radionuclides

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Abstract

Three different types of natural sorbents were developed using natural siliceous material, sodium alginate, and impregnation of goethite in alginate matrix. Experiments were carried out for decontamination of uranium, thorium, americium and plutonium by using siliceous material, Calcium-Alginate (Cal-Alg) and Goethite impregnated Calcium-Alginate (Geo-Cal-Alg) respectively. These materials were tested for different conditions by varying their pH in the range of 1-8, 95-98% Am-241 and Pu-239+240 sorbed by Cal-Alg and Geo-Cal-Alg beads respectively and experimental sorption capacity as evaluated were Eu^{3+} as a natural analogue for Am-241 and Pu-239+240 is 12.16 mg g^{-1}. Experimental results clearly showed that Th and U sorption capacity is 1 and 0.3 µg mg^{-1} (w/w) respectively in case of siliceous material. No change in various physicochemical characteristics of the potable water was observed after decontamination process.

Keywords: Natural sorbent; Siliceous material; Sodium alginate; Uranium; Thorium; Plutonium; Americium

Introduction

Drinking water contamination caused by Catastrophic incident like Dai ichi Nuclear Power Plant (NPP) Fukushima, Japan (11 March, 2011) and nuclear accidents like Chernobyl, USSR (March, 1986) demand scientists to evolve methodology for the decontamination of water without compromising on the drinking water quality. As per the reported literature data, from few Bq L^{-1} to 100 BqL^{-1} (approximately) of activity was reported in various drinking water sources in the region around accidental site [1-4].

In 2008, WHO published a third edition of the Guidelines for Drinking Water Quality [5]. 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 [5]. 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 activity respectively have been retained from the first edition. If activity of drinking water is below this level, no further action is required [5,6]. Allowable concentration in case individual alpha emitting radionuclides like Th-232, Pu-239+240 and Am-241 in drinking water is 1 Bq L^{-1} as per the guidelines of WHO issued in 2008. This value is 10 times higher comparing to recommended gross alpha value. Limit for uranium in drinking water is 15 µg L^{-1}, this is based on its chemical toxicity for the kidney instead of radiological toxicity. Details of the same are discussed elsewhere [7]. Thorium has number of isotopes in groundwater originating from both U-Th decay series. Under chemical equilibrium condition, ^{232}Th determines the chemical fate and speciation of all Th isotopes. A number of other Th isotopes (e.g., ^{234}Th, ^{230}Th, ^{228}Th) also exist in seawater, albeit at lower

concentrations, e.g., ^{234}Th is present in the ocean at atto-molar (10^{-18} M) concentration levels (Baskaran et al.). There is no specific limit for Th-232 in drinking water is given. In view of this; general guideline for gross alpha activity is taken into consideration. Therefore maximum allowable concentration in drinking water is 0.1 Bq L^{-1} [5]. Besides accidental scenario, radioactive constituents of drinking-water can result from 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), Radionuclide discharged from nuclear fuel cycle facilities. Manufactured radionuclides (produced and used in unsealed form), and past releases of radionuclides into the environment, including water sources.

Siliceous material was derived from soil. Solid Phases in soils is dominated by siliceous material. Ninty percent of solid phase is inorganic and made up of primary minerals (typically inherited from geologic material) like quartz, feldspar and mica and secondary minerals (these minerals result from weathering transformations of primary) like Vermiculite and Semectite [8,9].

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. The sodium alginate is generally used in various food items like jelly and hence its negative impact on the water quality is minimum. 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 have already been conducted to exhibit excellent adsorption ability for multivalent metal ions by calcium alginate beads [8,10,11]. 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 heavy metals such as Co, Cu, Cd and Zn by alginic acid [12-15] (Konishi et al., Min et al., Jang et al., Nayak et al.). 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 this polymer gel from potable water. The specificity of the alginate matrix for Pu-239+240 is enhanced by impregnating 2% geothite [α FeO(OH)] in the sodium alginate [11].

In the present study, decontamination of potable water from Uranium (U-238, 234, 235), Thorium (Th-232), Plutonium (Pu-239+240 and Amricium (Am-241) was carried out without disturbing the water qualities using natural sorbent followed by the treatment with activated charcoal column. Major water quality parameters were monitored to ensure the water quality.

Material and Methods

Spiking of the groundwater with U, Th, Pu and Am

Potable groundwater samples have been collected from three different sources (groundwater, rainwater and lake water) spread over a radial distance of 4 km from Trombay, Mumbai having latitude 19o00'498(N) and longitude 72o 55'136. Samples were filtered through 0.45 μm cellulose acetate membrane by using suction filtration. The stock solution of Pu-239+240 and Am-241 in 0.2 M HNO_3 and spectroscopy grade grade $Th(NO_3)_4$ and $UO_2(NO_3)_2$ solution of 1g L^{-1} were used to spike the filtered groundwater at pH 4.0 ± 0.3. pH of the solution was controlled by using 0.1 N $NaOH/HNO_3$. Various physicochemical parameters were measured before and after spiking the groundwater.

Preparation of siliceous material

Siliceous material was prepared by treating the soil particles (passed through to 2.0 mm sieve) with HNO_3 and H_2O_2 for six hours on the hot plate under reflux conditions. Siliceous particle were decanted and washed with demineralised water until completely neutralized and dried in the oven at 80°C. Details of the same were discuss elsewhere [9].

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. Goethite impregnated beads of Calcium Alginate was synthesized by mixing 2% (W/W) goethite in 2% solution of sodium alginate. Rest of the procedure is same as discussed above.

Synthesis of goethite

Synthesis of goethite was carried out at two different experimental conditions. In each case, 200 mL of suspension were prepared by mixing 100 mL of $FeSO_4.7H_2O$ solution with 100 mL of NaOH solution. The rotation speed of the magnetic stirrer was set at 760 rpm. The first experimental reference is defined by the following conditions. The reaction was carried out at room temperature (T= 25°C) keeping the ratio (R) of [$FeSO_4$]/[NaOH] as 0.2 [$FeSO_4.7H_2O$=0.15 mol L^{-1}; NaOH=0.75 mol L^{-1}]. Whereas in the second case R was 2.5 and the concentrations of $FeSO_4.7H_2O$ and NaOH was 0.5 mol L^{-1} and 0.20 mol L^{-1} respectively. With experimental condition (I), goethite forms directly from $Fe(OH)_2$. With experimental condition (II), goethite forms from an intermediate Fe(II–III) compound [16,11].

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

Various physiochemical characteristics like pH, conductivity and redox potential was 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 was either measured by Flame Atomic Absorption Spectrometry (FAAS) 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.

Measurement of Pu-239+240 and Am-241

The measurement of alpha activity of Pu-239+240 and 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, detailed of the same discussed elsewhere [6]. This electrodeposited source was calibrated against mixed nuclide source Am-241 from Amersham (AMR 33).

Measurement of U and Th

Determination of uranium and thorium was carried out by using simultaneous solid state detector Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES, model ACTIVA, from Horiba Jobin Yvon SAS, France). Intensity of emission was measured at two different wavelengths i.e. 398.582 and 367.011 nm in the case of uranium whereas thorium emission was measured at 401.913 and 283.232 nm. The calibration of the instrument was carried out by using Aldrich standard solution of uranyl nitrate and thorium nitrate. ACTIVA utilizes a 2048x512 pixels, ultra-low noise and high quantum efficiency Charge Coupled Device (CCD) solid-state detector. ACTIVA includes a unique optical design featuring a 0.64m Czerny-Turner optical system and holographic gratings of 4343 grooves per mm and 2400 grooves per mm, covering the full 6 mm height of the plasma; "Normal Analytical Zone Multi-WAV" acquisition mode provides complete sample fingerprinting of 75 elements in less than 30 seconds. It gives resolution up to 10 pm. Concentrations of uranium and thorium in different samples was also validated by using alpha spectrometry.

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}.

Characterization of goethite

Malvern, Zeta-sizer nano zs with MPT-2 auto-titrator was used for the measurement of hydrodynamic diameter of the goethite suspension. This technique is based on Dynamic Light Scattering (DLS) and measure the hydrodynamic diameter in the range of size 0.6 nm-6 μm, 1.5 mL of the aliquots were taken in the transparent cell. Goethite characterization in dry stage was done by PXRD (Powder X-ray Diffraction) and ATRFTIR (Attenuated Total Reflectance Fourier Transform Infrared) Spectroscopy. SEM-EDS (Scanning Electron Microscope with energy dispersive spectrometry) were used for morphology and elemental characterization of goethite. Siliceous material/Calcium alginate beads/goethite impregnated calcium alginate & granular activated charcoal column.

Siliceous material/Calcium alginate beads/goethite impregnated calcium alginate and granulated activated charcoal column

It was prepared by putting 20 g (wet weight) of each sorbent and 10 g of granulated activated charcoal separately in glass columns (20 cm long and 0.8 cm diameter). 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 liter) on top. Before elution from the natural sorbent column, holdup time of 30 and 100 minutes was given in case of Th and U respectively. The eluted solutions from the natural sorbent columns were passed through activated charcoal column. The rate of elution was maintained at a speed of 2 mL min^{-1} in both the column.

Results and Discussion

The important water quality parameters monitored for the potable water collected from three different sources (groundwater, rainwater, lakewater) and 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 TDS but still much below than the permissible limit [5].

Parameters	Average Value (μg L^{-1})		
	Ground water	Rain water	Lake Water
pH	7.7 ± 0.32	4.2 ± 0.21	5.1 ± 0.19
Conductance (μS)	280 ± 10	120 ± 10	100 ± 16
Total hardness	120 ± 4	65 ± 2.7	70 ± 3.2
HCO_3^-	65 ± 3	25 ± 2	32 ± 1.3
Dissolved Organic Carbon (DOC)	0.8 ± 0.02	0.3 ± 0.08	1.30 ± 0.08
Total Dissolved Solid	300 ± 20	150 ± 10	100 ± 5
Turbidity (NTU)	6 ± 0.4	2 ± 0.1	4 ± 0.3
PO_4^{3-}	0.35 ± 0.02	0.15 ± 0.02	0.32 ± 0.03
NO_3^-	4.5 ± 0.8	1.2 ± 0.5	1.8 ± 0.2
F^{-1}	0.07 ± 0.01	0.03 ± 0.01	0.02 ± 0.01
Calcium	24.23 ± 1.20	12 ± 0.4	14 ± 1.3
Magnesium	7.4 ± 1.2	5.6 ± 1.1	6.7 ± 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

Table 1: Characteristics of potable water collected from different sources

Formation of calcium alginate beads

Calcium alginate beads formation from solution of sodium alginate involves a simple displacement reaction in which sodium ions are replaced by Ca^{2+} These beads have enough mechanical strength and were stable for 3- 4 weeks. Details of the same were discussed elsewhere [7,9]

Characterization of siliceous material and calcium-alginate (Cal-Alg) and goethite-calcium-alginate (Goe-Cal-Alg) beads

Detailed characterization of natural sorbent developed for this work was done by using ATR-FTIR, PXRD, SEM-EDS. Details of the same were discussed elsewhere [7,9,11,17,18].

pH optimization for sorption of Am-241 and Pu-239+240 and by calcium alginate beads (Cal-Alg) and goethite impregnated calcium alginate beads (Goe-Cal-Alg) column

In order to have effective sorption of Am-241 and Pu-239+240 by the Cal-Alg and Goe-Cal-Alg beads column respectively, pH is optimized. Figure 1 shows the sorption profile of Am-241 and Pu-239+240 having a concentration of 500 Bq L^{-1} at different pH.

From the Figure 1 it is clear that at pH 4 more than 99% Am-241 and Pu-239+240 is sorbed by Cal-Alg and Goe-Cal-Alg beads respectively. This is attributed due to the ionization of COOH group of the alginate biopolymers and goethite was also partially ionized to FeO^- and OH^- [19,20]. At this pH maximum ionization is achieved and these ionizable sites are available for binding with Am^{3+} and Pu^{4+} At pH greater than 4, due to higher concentration of OH^- ions the Am is present as neutral species $Am(OH)_3$ and Pu as $Pu(OH)_4$ with a small fraction as $Pu(OH)_6$ and not available to bind with COO^- and FeO^-.
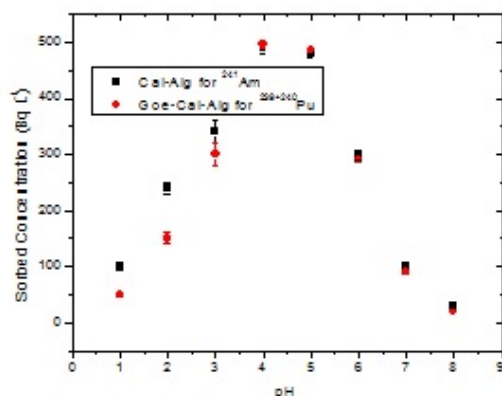
Figure 1: Variation in sorption of Am-241 and Pu-239+240 on Cal-Alg and Goe-cal-Alg column respectively with pH

Optimization of pH for the sorption of U & Th on siliceous material

Sorption of ions to particle surfaces is strongly pH dependent, with cations generally showing increasing sorption with increasing pH, and anions showing increasing sorption with decreasing pH. In order to see the effects of pH on the sorption of U and Th on siliceous matter experiments were carried out in the pH range of 1-6, keeping the constant concentration of Th/U (10 μg mL^{-1}) and siliceous material (500 mg). The maximum sorption was observed at pH 4 for both U and Th. However in case of uranium maximum sorption is only 70% whereas for thorium it is 94% (Figure 2).
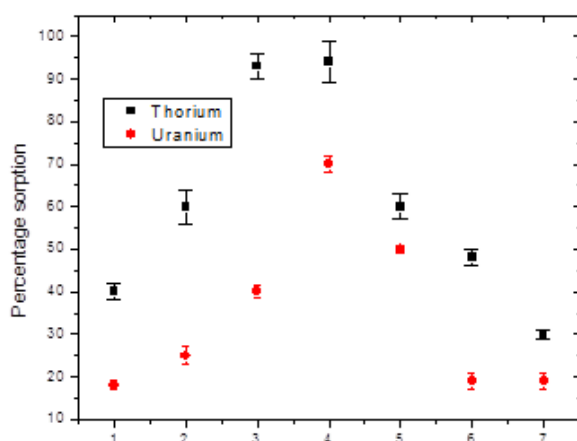

Figure 2: Variation in sorption of uranium and thorium on siliceous material column with pH

Siliceous materials were developed by destruction of organic layers from soil particles therefore elevated level of thorium sorption

compare to uranium is mainly due to its particle reactive nature. As other major complexing ligands such as humic/fulvic acid are not present on the surface of soil therefore only driving force for the sorption of U and Th is "particles surface activity". This particle reactive driving force favor thorium due to its single predominant oxidation state (IV) leading to constant ionic potential. Enrichment of Th(IV) in marine sediment having more than 70 % siliceous matter was also reported by Basu et al. [17].

Variation of sorption of Am-241, Pu-239+240, U and Th by Cal-Alg, Geo-Cal-Alg beads and siliceous material respectively with time

Figure 3 gives variation of sorption of Am-241 and Pu-239+240 with time whereas the same in case of U & Th by siliceous material is shown in Figure 4. From these figures, it is evident that equilibrium sorption time in case of Am and Pu-239+240 by Cal-Alg and Geo-Cal-Alg beads is 100 minutes whereas the same in case of siliceous material is 30 minutes for Th and 100 minutes for uranium. In view of this, elution of the spiked samples in all the experiment was done after reaching the equilibration time, maintaining the elution rate of 2 mL m^{-1}.
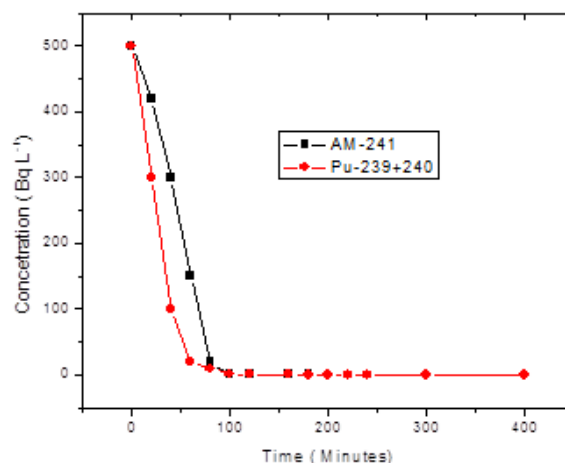

Figure 3: Variation in sorption of Am-241 and pu-239+240 on siliceous material column with time

Evaluation of the sorption capacity of the column for Am-241 and Pu-239+240

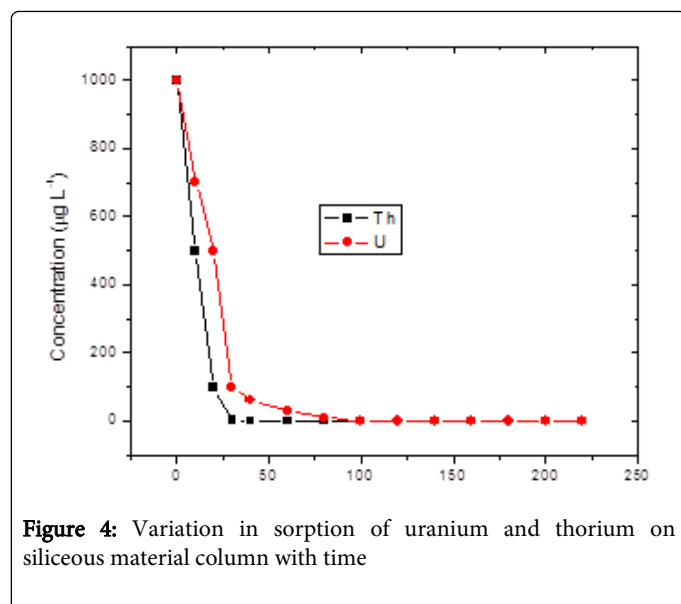To meet the radiological regulatory requirement of the lab, sorption capacity was evaluated with Eu^{3+}.

Figure 4: Variation in sorption of uranium and thorium on siliceous material column with time

Since the amount of actinides (Pu, Am) in environmental sample is at ultra-trace level. In view this europium (Eu) was used as natural analog for Pu & Am. Using Eu is necessary when macro amounts are needed for evaluation of sorption capacity. Sorption isotherms of Eu^{3+} for alginate gels were obtained in a wide range of initial concentration of Eu^{3+} from 1.3×10^{-4} to 6.6×10^{-2} M. The stability of the higher oxidation states of the transuranium elements typically europium is the heavy homolog of americium and plutonium decreases with increasing atomic number [10,19,20,21]. In agreement with this, the trivalent state of americium is the most stable oxidation state, and it is the homolog of americium, [Am^{3+} and Pu^{4+} radius=0.99 AO and 1 AO respectively [22]. The equilibrium amount of Eu^{3+} adsorbed on alginate gel increased with equilibrium concentration of Eu^{3+}, and then approached a constant value at Eu^{3+} concentration. Experimentally observed sorption capacity is 12.16 mg g^{-1} which is very close to the Langumir constant Q_0 (see Figure 5),
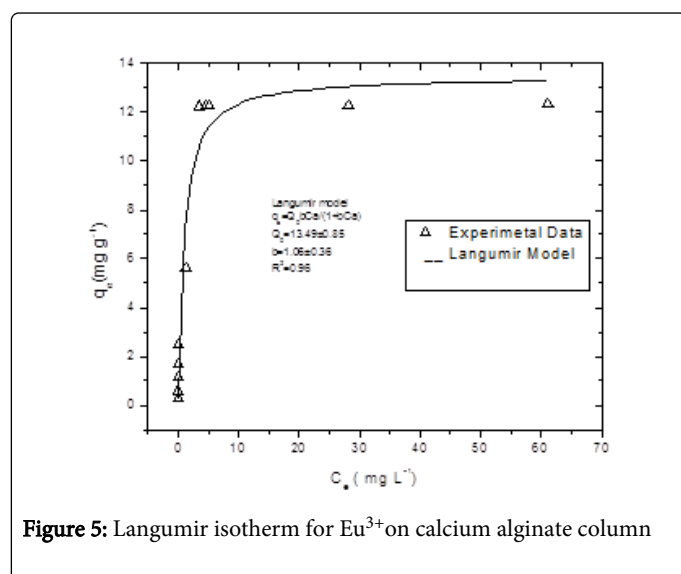

Figure 5: Langumir isotherm for Eu^{3+} on calcium alginate column

Which is measure of the monolayer adsorption capacity, as obtained, 13.49 ± 0.85 mg g^{-1} . Figure 6, gives the experimental fit of

Freundlich isotherm model where constant k and n are related to the strength of the adsorptive bond and the bond distribution. This model assumes that the ratio of the amount of solute adsorbed on to a given mass of adsorbent to the concentration of the solute in the solution is not constant at different concentrations. As clear from the Figure 5 and 6 experimental data are very close to the Langmuir-type adsorption model.
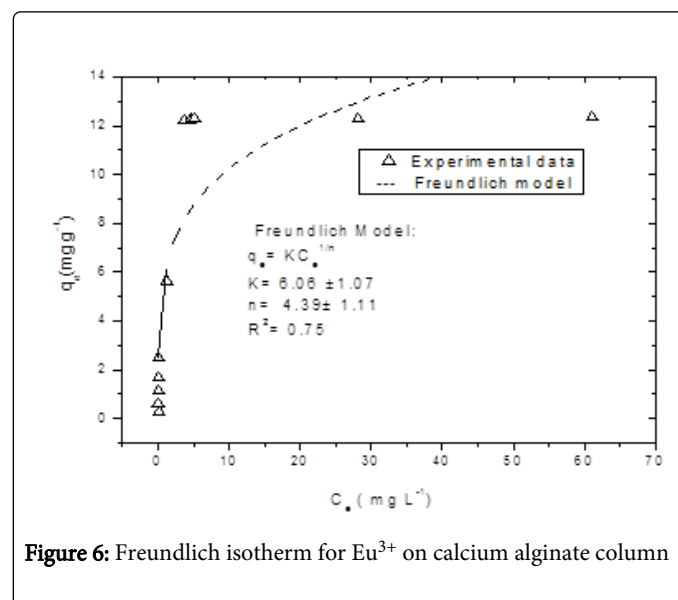

Figure 6: Freundlich isotherm for Eu^{3+} on calcium alginate column

Sorption of U and Th on siliceous material

Figure 7, gives the variation in percentage sorption of U and Th keeping the concentration of adsorbate same (50 mg) while the concentration of U and Th vary between 1 µg mL^{-1} to 10 µg mL^{-1} (Total volume 50 mL). From this Figure it is clear that uniform sorption (µg mg^{-1}) was observed irrespective of Th/U concentration. Experiments clearly show that the sorption capacity of silicious material for thorium and uranium was 1 and 0.3 µg mg^{-1} respectively for different concentrations of thorium and uranium. From these experimental results it is clear that in the absence of any complexing ligand such as humic/fulvic acid on the surface of soil "particle's surface activity" is the only driving force to sorb Th and U on the siliceous material. In order to understand the various species of uranium present at pH 3-4, work carried out by [23] using computer program EQ3/6 was used. As per this uranium speciation model at 4×10^{-4} M initial uranium solutions showed that the uncomplexed uranyl ion UO_2^{2+} dominates the U(VI) speciation at pH 3-4 and with the increase of pH, fraction of UO_2^{2+} decreases sharply [24-28].
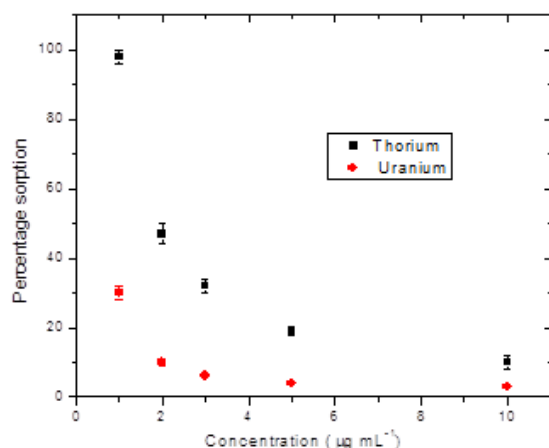
Figure 7: Variation in sorption of uranium and thorium on siliceous material column at different concentration of uranium and thorium

Whereas in case of Thorium, no change in sorption was observed with a change in pH. This is because chemical properties of Th are relatively simple. Th has only one stable oxidation state, IV, under all redox conditions in natural waters, and Th(IV) ions are extremely particle reactive. The reactions and equilibrium equations for sorption of Th on silica are very similar to the proton exchange [29-34].

Parameters		Average Value ($\mu g\ L^{-1}$)					
		Cal-Alg column		Goethite–Cal-Alg. Column		Siliceous matter Column	
After Passing Through		Calcium Alginate column	Activated Charcoal Column	Calcium Alginate-Goethite column	Activated Charcoal Column	Silicious matter column	Activated Charcoal Column
pH		4.2 ± 0.3	4.1 ± 0.4	4.3 ± 0.3	4.4 ± 0.3	5.0 ± 0.4	5.0 ± 0.4
Conductance (µS)		230 ± 15	220 ± 9	125 ± 8	132 ± 11	100 ± 7	110 ± 5
Total hardness		70 ± 10	65 ± 10	78 ± 8.7	55 ± 8.7	62 ± 6.2	62 ± 6.2
HCO_3^-		55 ± 6	55 ± 6	35 ± 1.3	35 ± 1.3	30 ± 1.3	30 ± 1.3
Dissolved Organic Carbon (DOC)		1.0 ± 0.02	1.0 ± 0.02	0.8±0.08	0.8±0.08	1.30±0.12	1.30±0.12
Total Dissolved Solid		260 ± 10	250 ± 15	140 ± 7	145 ± 8	110 ± 12	123 ± 5
Turbidity (NTU)		3 ± 0.2	3 ± 0.4	3 ± 0.2	3 ± 0.1	5 ± 0.1	5 ± 0.12
Ca^{2+}		46.16 ± 3.45	19.6 ± 3.20	41 ± 1.2	17 ± 1.7	15 ± 1.5	14 ± 1.4
NO_3^-		6.0 ± 0.3	4 ± 0.5	8.2 ± 0.2	2.2 ± 0.8	3.6 ± 0.4	3.6 ± 0.4
Cl^-		280	250	400	350	450	380
Na^+		$440x10^3$	$280x10^3$	$360x10^3$	$280x10^3$	$340x10^3$	$285x10^3$
DO (Dissolved Oxygen)		6.9 ± 1.6	6.5 ± 1.4	6.9 ± 1.3	7.1 ± 1.3	6.8 ± 1.3	6.5 ± 1.3
Gross Alpha ($Bq\ L^{-1}$)		<0.1	<0.1	<0.1	<0.1	<0.1	<0.1
Gross Beta ($Bq\ L^{-1}$)		<1.1	<1.1	<1.1	<1.1	<1.1	<1.1

Table 2: Characteristics of the water samples after decontamination (After passing through the natural sorbent column i.e. column of either of Cal-Alg; Goe-Cal-Alg; Silicious material) and granulated activated charcoal column

The purification of process water from natural sorbent using activated charcoal column

After having decontamination of U, Th, Pu and Am from the water samples by using natural sorbent column, solution were further passed through the charcoal column for purification of potable water. Table 2 gives the values obtained for various parameters. From the data it is clear that the parameters (Ca^{2+}, NO_3^{-1}, Na^+ and Cl^-) are not much different from the original parameters given in the Table 1. Small variation in the parameters in both the cases may be due to the absorption of some of the metal ions at the charcoal column.

Conclusion

Low level of U, Th, Pu and Am were sorbed from potable water without disturbing the various water quality parameters by using natural sorbents (Siliceous matter, Cal-Alg and Goe-Cal-Alg). Control on the concentration of Ca^{2+}, NO_3^{-1}, Na^+ and Cl^- was achieved by using activated charcoal column in conjugation with natural sorbent.

Acknowledgement

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