



CHEMICAL EFFECTS OF GOETHITE COLLOID ON THE TRANSPORT OF URANIUM (VI) THROUGH A SATURATED QUARTZ-PACKED COLUMN

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Abstract

Laboratory experiments were carried out to determine the breakthrough of uranium (VI) through a saturated porous wedron-510 sand column in absence and presence of goethite colloid (α -FeOOH). NaCl solution in the ionic strength range of 10^{-4} M- 10^{-2} M at pHs 7.5 and 10.5 and Inshas groundwater were used to saturate wedron-510 sand column. In the presence of CaCO_3 , the species of U(VI) were UO_2^{2+} , $(\text{UO}_2)_2\text{CO}_3(\text{OH})_3^-$, $\text{UO}_2(\text{OH})_3^-$, and $\text{UO}_2(\text{OH})_4^{2-}$. At pH 7.5, the transport of U(VI) at 10^{-4} M NaCl was retarded, while it facilitated at 10^{-3} and 10^{-2} M NaCl in the presence of goethite colloid. On the other hand, at pH 10.5 the transport of U(VI) was facilitated in the presence of goethite colloid. In Inshas groundwater the transport of U(VI) was retarded in the presence of goethite colloid.

1. INTRODUCTION

Synthetic goethite (α -FeOOH) is important in waste management as well as in industry. In waste management, it could affect the transport of radionuclids in unsaturated and saturated (groundwater) zones. In nature goethite could be initiated as a result of corrosion of steel waste drums, engineering barrier reinforcement steel or as impurities in the soil around the disposal sites. Colloidal particles are potential vehicles for pollutants, migrating rapidly through the medium because of their average velocity within the solid medium which is somewhat greater than the solute average velocity. It has been reported that mobile subsurface colloids can carry groundwater contaminants adsorbed onto their surfaces [1]. In liquid waste disposal projects, groundwater quality can be endangered by suspended clay and silt particles migrating from the formation adjacent to the well bore. McCarthy and Zachara [2] noted that failure to account for the role of colloids in facilitating contaminant transport can lead to serious underestimation of the distances the groundwater contaminants can migrate. Puls and Powell [3] studied some parameters such as flow rate, pH, ionic strength, electrolyte composition, and colloid size, in studying arsenate transport in the presence of Fe_2O_3 particles in a sand and gravel column. The colloid breakthrough was highly affected by its particle size and anionic composition of the electrolyte. Satmark et al.[4] studied the chemical effects of goethite colloid on the transport of ^{137}Cs , ^{22}Na , and ^{131}I through a quartz-packed column. They concluded that pH is a controlling parameter for goethite transport. At pH 4, goethite is strongly retained by the quartz and the deposition seems to be irreversible. A small alteration of the breakthrough curve for cesium was noticed compared to the tritiated water at pH 4 and 10.5. No such effect was seen for sodium at these pHs. Goethite has no effect on the transport of Na at pH 10.5.

In this work, the transport of $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, uranyl nitrate hydrate (UNH) was studied in the presence and absence of goethite colloid. All the experiments were conducted at room temperature ($25 \pm 2^\circ\text{C}$) using NaCl solution and Inshas groundwater..

2. MATERIALS AND METHODS

2.1 Synthesis of Goethite Colloid Particles(α -FeOOH)

All the chemicals used in this work are Analar Grade Reagent. Solutions were prepared using double distilled water (DW) from a Milli-Q plus water purification system which had a resistivity of 18.0 M Ω .cm and a pH range of 3.74-4.0. Goethite colloid particles were prepared according to the method of Atkinson et al.[5], using Fe(NO₃)₃. The pH of the Fe(NO₃)₃ solution was rapidly raised to pH 11.8-12.0 by addition of NaOH and then hydrated and aged for 5 days at a 63°C temperature oven to allow conversion of Fe(NO₃)₃ to a mixed-valence, then crystallized to precipitate α -FeOOH [1]. The surface area using N₂ – BET adsorption method and density were determined to be 51.8 m²/g and 4.81 cm³/g respectively. Wedron-510 is a commercial silica sand, mainly quartz [1] purchased from Wedron Silica, USA. The ²³⁸U(VI) used in this study is in the form (UO₂(NO₃)₂.6H₂O), its molecular weight is 502.13. The UNH purchased from Fisher, USA.

2.2 Measuring Techniques

The U(VI) measurements made by the UA-3 Scientrex instrument are based on the fluorescence of a uranyl complex formed by addition of a reagent to the sample during analysis with a sensitivity better than 0.05 ppb of uranium, it was produced by Scientrex, Canada. Coulter multisizer II was used to determine the number of goethite colloidal particles, it was produced by coulter scientific instruments, USA.

2.3 Procedure

The transport of UO₂(NO₃)₂.6H₂O and UO₂(NO₃)₂.6H₂O / colloidal goethite in saturated porous medium experiments were conducted using a 2.5 cm diameter by 15.0 cm long Plexiglass column was filled with 130.0 g of wedron-510 sand and saturated with 18.1 g deionized distilled water (pore volume). To check the backing of the column with wedron-510 sand and the flow of chemical solution through the column, and the dispersion along the column and there is no channeling, clean bed breakthrough curve experiments were conducted. Potassium sulfate (K₂SO₄) tracer was used [1]. About 110 ppm SO₄ were pumped in the saturated wedron-510 sand column. The anion SO₄⁻ was used to obtain a continuous and an input pulse breakthrough curve. About 200 ml of NaCl contains 75 ppm CaCO₃ were pumped bottom up of the column.

The experimental setup is shown in Fig.1. One ml of 18 mM UO₂(NO₃)₂.6H₂O was added to the NaCl buffer solution. One ml of colloidal goethite containing about 5.0x10⁷ particles was added to the chemical solution in the case of studying the effect of goethite colloid on the transport of UO₂(NO₃)₂.6H₂O. The flow rate of pumping the solution was 1.0 ml/min giving 0.5 pore volume per 8 minutes. The ionic strength range of NaCl studied was 10⁻⁴-10⁻²M and the pH's were 7.5 and 10.5.

3. RESULTS AND DISCUSSIONS

It was found that the transport of U(VI) depends on the hydrology of the system, species of uranium (VI), the adsorption/desorption of U(VI) onto both the goethite and the wedron-510 sand surfaces [1].

Figure 2. shows that the major species of U(VI) are UO₂²⁺, (UO₂)₂CO₃(OH)₃⁻, UO₂(OH)₃⁻, and UO₂(OH)₄²⁻ using EQ3NR/EQ6 computer code [6]. The flow in the porous medium is described by Darcy's velocity [7]. The pore velocity is given by:

$$V = -\frac{k\rho g}{\omega\eta} \cdot \frac{dh}{dl} \quad (1)$$

where k is the permeability calculated to be (4.0 x 10⁻⁷ cm²), ρ is the water density (1000kg/m³),

g is the acceleration of gravity (9.8 m/s^2), h is the hydraulic head (m), l is the distance between two head points, η is the viscosity of the water at 25°C equals to $0.891 \times 10^{-3} \text{ kg/m.s}$, and ω is the porosity of the porous medium found to be 0.30 [1]. The quantity $\frac{k_{pg}}{\eta}$ is the hydraulic conductivity of the system found to be $4.4 \times 10^{-2} \text{ cm/s}$. The pore velocity (V) was found to be $1.45 \times 10^{-2} \text{ cm/s}$. Peclet and Reynold's number were found to be 2.92 and 1.03 respectively, which suggest that the flow in the system is laminar and dependent on the dispersion as well as the advection. Figure 3. shows that the transport of UNH was retarded in Inshas groundwater in the presence of goethite colloid. This could be attributed to the decrease the adsorption of UNH as a result of the competition with Na^+ , K^{2+} and Mg^{2+} cations present in the groundwater [1]. At pH 7.5, the transport of U(VI) at 10^{-4} M NaCl was retarded, while it facilitated at 10^{-3} and 10^{-2} M NaCl in the presence of goethite colloid (Fig.4.). On the other hand, at pH 10.5 the transport of U(VI) was facilitated in the presence of goethite colloid (Fig.5.). This could be attributed to the repulsion force between the U(VI) species (Fig.2.) and the negative charged surfaces of quartz and goethite at pH 10.5.

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TABLE I: COMPOSITION OF CHEMICAL SOLUTION

Species	Concentration mg/L
HCO_3^-	46.4
UO_2^{++}	22.69
NO_3^-	10.42
Na^+	23.0
Ca^{++}	30.4
Cl^-	35.0
pH	7.5 or 10.5

Table I: Composition of chemical solution

Captions of figures:

Fig.1. The experimental setup of breakthrough curves experiments.

Fig.2. Speciation of uranium (VI) /CaCO₃ as a function of pH at 10⁻² M NaCl solution at room temperature (25 ± 2 °C) .

Fig. 3. Effect of goethite colloid on the transport of UNH in saturated porous wedron-510 sand in Inshas groundwater compared with K₂SO₄ tracer (25 ± 2 °C).

Fig.4. Effect of goethit colloid on the transport of UNH in saturated porous wedron-510 sand at different ionic strengths at room temperature (25 ± 2 °C) at pH 7.5.

Fig.5. Effect of goethit colloid on the transport of UNH in saturated porous wedron-510 sand at different ionic strengths at room temperature (25 ± 2 °C) at pH 10.5.

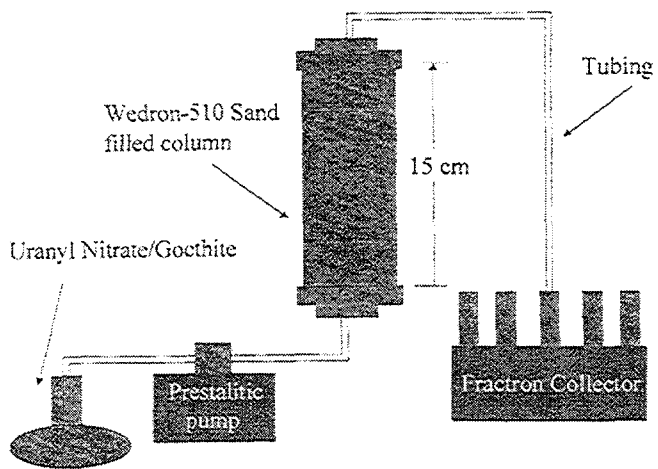


Fig.1.

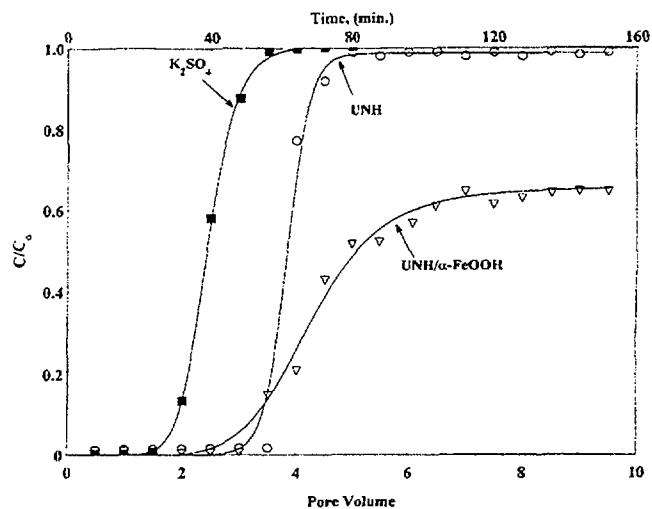


Fig.2.

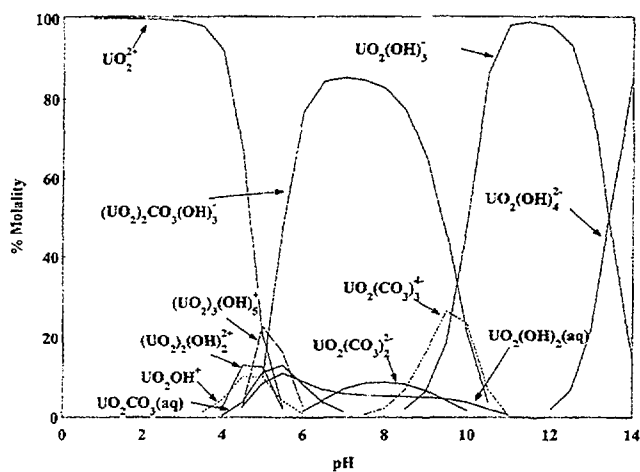


Fig.3.

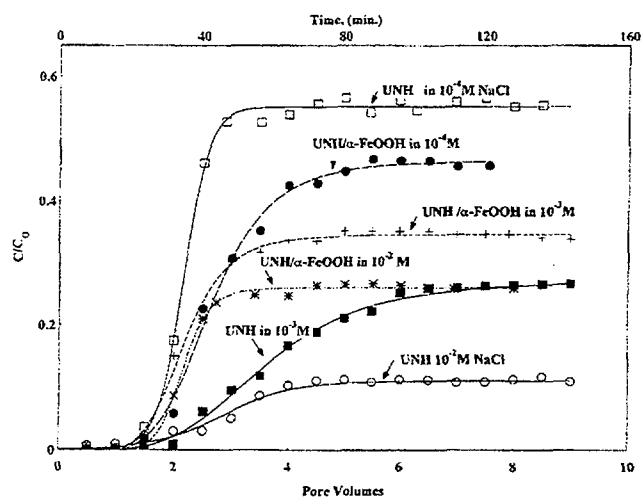


Fig.4.

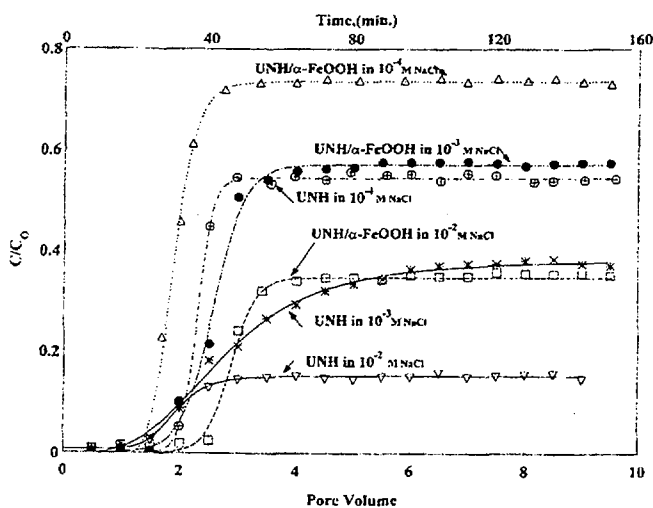


Fig.5.