Separation Science and Technology
Publication details, including instructions for authors and subscription information:
http://www.informaworld.com/smpp/title~content=t713708471

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To cite this Article Fondeur, F. F. , Hobbs, D. T. and Fink, S. D.(2010) 'The Effect of Magnetic Fields on Uranium and Strontium Sorption on Monosodium Titanate', Separation Science and Technology, 45: 12, 1876 — 1879
To link to this Article: DOI: 10.1080/01496395.2010.493107
URL: http://dx.doi.org/10.1080/01496395.2010.493107

Separation Science and Technology, 45: 1876–1879, 2010

ISSN: 0149-6395 print/1520-5754 online
DOI: 10.1080/01496395.2010.493107

The Effect of Magnetic Fields on Uranium and Strontium Sorption on Monosodium Titanate

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The presence of a magnetic field gradient enhanced the rate of strontium and uranium sorption onto monosodium titanate. The enhancement was evident only in the early contact times and did not impact the equilibrium capacity of the sorbent. No enhancement was seen when the sorption test was conducted under a homogeneous magnetic field. Further studies are needed to determine if the enhancement is also seen with other cations and/or anions.

Keywords actinide; gradient field

INTRODUCTION

Adulterated water (accidental or intentional contamination) is threatening the amount of potable water available to life. The rate of water contamination throughout the world is far outpacing efforts to restore water purity. To limit the extent of contamination, several technologies are deployed to purify water (1). These technologies include ion exchange, precipitation, distillation, sorption, solvent extraction, filtration, and many hybrid technologies that incorporate organic destruction processes. Solute separation from aqueous solvent is also a key pretreatment step in the safe immobilization of liquid waste especially nuclear waste where some solutes are toxic and radioactive.

The Savannah River Site (SRS) conducts separation treatment operations of radioactive supernate with a monosodium titanate, $NaHTi_2O_5 \times H_2O$ (MST) sorbent (2). This sorbent selectively removes strontium and actinides from strongly alkaline salt solutions. The rate of removal depends on the type of ion in solution. Strontium sorbs very rapidly and typically equilibrium conditions are achieved within 24 hours. Sorption of the actinide ions occurs more slowly and can take several days to weeks to reach equilibrium conditions. Thus, the removal of the actinides determines throughput in processing facilities.

Various methods have been considered to accelerate the sorption rate of actinides onto MST.

These methods include

1. chemical modification of MST to yield higher surface area and ionic diffusivity,
2. intensification (or optimization) of the batch processing parameters such as mixing power and residence time, and
3. use of force fields to enhance ionic transport or sorption.

Chemical modification of MST using hydrogen peroxide has shown promise as the modified MST (mMST) exhibits faster uptake of strontium and plutonium. The enhanced adsorption rates of the mMST is attributed to increased surface area and porosity (3–5). Increased mixing energy or increased residence time may lead to increased removal of the target radionuclides, but the added energy increases cost and the increased residence time decreases the throughput of operations.

As a third method to accelerate processing, we considered applying an alternating current (AC) electric field to enhance sorption process. Tests indicated that the alternating of an applied AC electric field heats the MST (resulting from friction of the alternating dipoles in MST) and enhances ionic diffusivity within MST. Despite the improvement, the added energy cost and added safety precaution of creating and maintaining an AC field (AC oscillator) are significant concerns of this approach. We next considered using a permanent magnetic field to increase sorption under the assumption that a magnetic field may disrupt the hydrogen bonding network of water inside the MST pores. Most oxide surfaces are hydrated in solution and have a few layers of sorbed water (forming an inner sphere). For an ion to absorb on the oxide surface, the sorbing ion must push this water layer away from the oxide surface. Permanent and variable magnetic fields are currently used to separate ferrous and paramagnetic materials from solid mixtures (6). Similarly, suspensions of paramagnetic particles are separated from solutions using magnetic field gradients generated from suspended fibers. Our objective in this project was to determine if

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Received 1 November 2009; accepted 12 March 2010.

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the application of a gradient or continuous magnetic field affected strontium and actinide sorption.

EXPERIMENTAL

A salt solution with the composition listed in Table 1 was prepared to simulate the composition of a radioactive liquid wastes at the Savannah River Site (SRS). All the chemicals used for the salt simulant preparation were ACS reagent grade. The water used for this simulant was doubled distilled and de-ionized water. This solution was spiked with trace amounts of ^{85}Sr for tracking the concentration of strontium. To this solution monosodium titanate was added to generate a slurry of 0.8 grams of MST per liter of solution. The slurry spun in 5 mL polybottle (polypropylene) at 30 and 300 rpm (0.026 m/s and 0.26 m/s respectively). The spinner is a Lightnin Labmasters model L5U10 from SPX Process Equipment, Rochester, NY. The spun solution was subjected to a homogeneous magnetic field (0.9 Tesla) and to a gradient magnetic field (39.7 T/m). For comparison, baseline tests were conducted under no magnetic field (at the same time) with identical mixing. Figure 1 is a picture of the mixing test. Also shown in Fig. 1 are the two different magnetic geometries (NdFeB) that generated the homogeneous magnetic field and the magnetic field gradient.

Samples were spun under the magnetic field at different length of times (2, 4, 18, and 68 hours). At the end of the mixing time, the slurry was filtered with 0.1 micron nylon filter attached to a syringe where a plunger forced the slurry through the filter. The collected filtrate was analyzed by ICP-MS for uranium (^{238}U) and strontium (^{85}Sr).

TABLE 1
The composition of the salt simulant tested

Component	Concentration	Susceptibility*
Free OH (M)	1.38	1.2E-11
NO_3 (M)	3.11	−2.3E-11
NO_2 (M)	0.16	−1.50E-11
SO_4 (M)	0.61	−4E-11
Al (M)	0.46	−3.1E-12
Calculated CO_3 (M)	0.039	−3.4E-11
Calculated Total Na (M)	6.39	−2.3E-12
Water (M)	43	−1.6E-10
^{87}Sr (μg/L)	543	−9.0E-12
^{238}U (μg/L)	9.87 E 3	−2.0E-07

Total solution susceptibility = −7.07E-9 (gram/gram).
*From Reference 7.

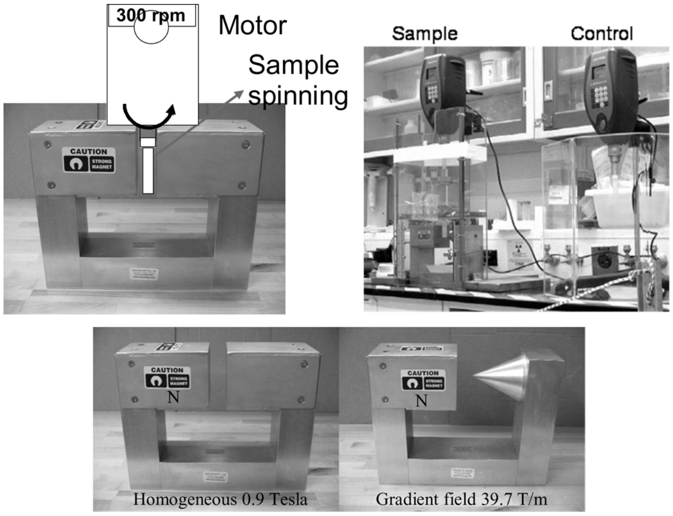

FIG. 1. A picture of the magnet geometry (NdFeB) and the location of the sample vial within the magnet gap.

RESULTS

We conducted a long term (72 h) kinetics test under a magnetic field to determine any long-term effect. Figure 2 shows the amount of uranium on MST (μmole/gram of MST) as a function of time (hours) while exposed to 0.8 g/L MST for three days. Also shown in Fig. 2 is the uranium data from the same test under no magnetic field. The two sets of data (with and without magnetic field) reached the same equilibrium value (i.e., same loaded uranium concentration). The two sets of data numerically differed early (at 2 hours contact time) in the test outside the statistical uncertainty. This difference is due to the presence of a magnetic field gradient. The error bars in this and other figures reflect the 95% confidence interval value of the triplicate experiments (except for the right figure in Fig. 3 where duplicates were conducted).

Given the above result, testing focused on examining magnetic effects on sorption at the first two hours of contact with MST. Figure 3 shows the effect of a gradient magnetic field on the strontium and uranium loading on MST after two hours of contact. Visual inspection of Fig. 3 clearly shows that the presence of a magnetic field gradient increased the Sr and U loading by a factor of three (in the case of Sr) and 2.5 (in the case of U), respectively, relative to the loading test under no magnetic field.

Figure 4 shows the effect of the magnetic field type (homogeneous versus a gradient) on the uranium loading on MST at high Reynolds numbers (2000), or under turbulent conditions. Sorption under a gradient magnetic field was nearly 1.5 times larger than under a homogeneous magnetic field. Sorption under a homogeneous magnetic field was slightly lower than that with no magnetic field. Thus, the type of magnetic field (gradient) positively affects

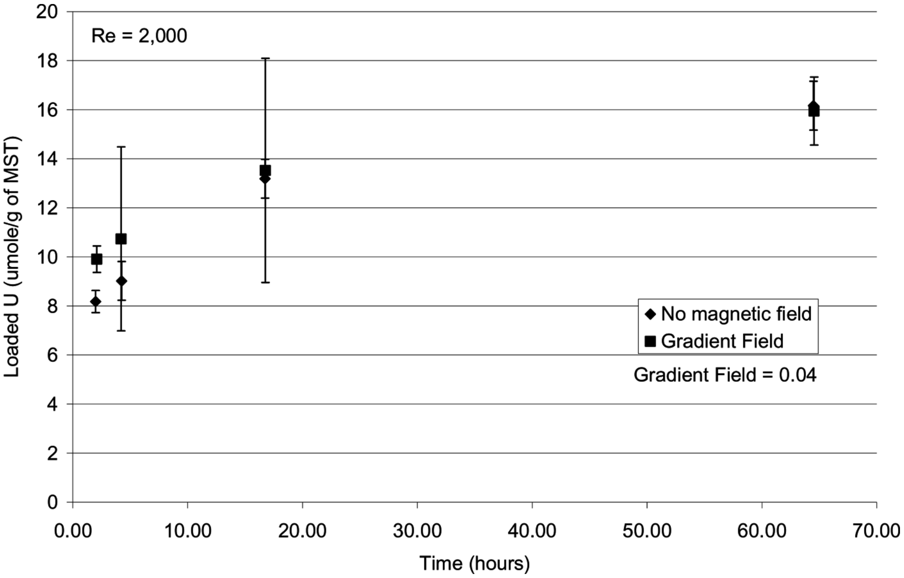

FIG. 2. Data obtained at two hours indicate a faster uranium loading. The samples were stirred at 300 rpm (high speed). The convergence of the data at later time is an indication of the same capacity. All strontium data measured below detection limits (at 300 rpm mixing rate). The error bars reflect a 95% confidence interval. The error bars at two hours are smaller than the data pattern.

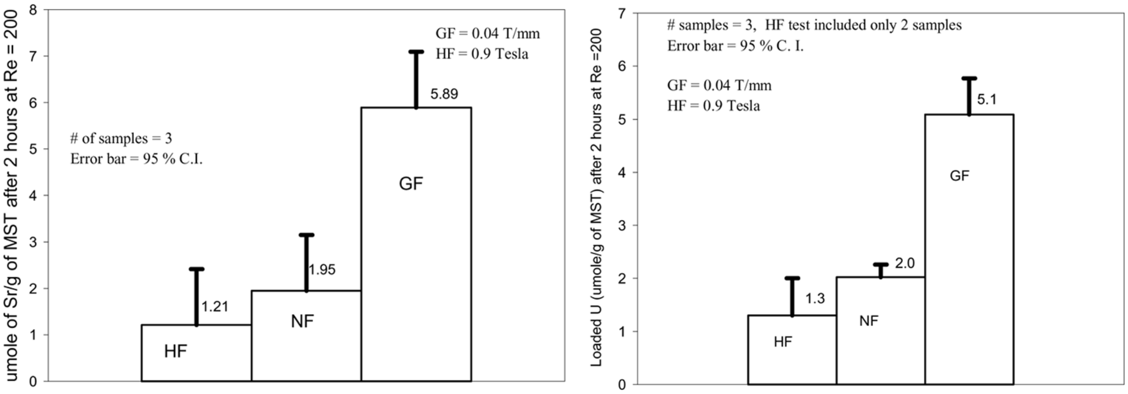

FIG. 3. More strontium and uranium loaded on MST under a magnetic field gradient after two hours of exposure to MST stirred at 30 rpm (low speed). NF = no field, HF = homogeneous field, and GF = gradient field.

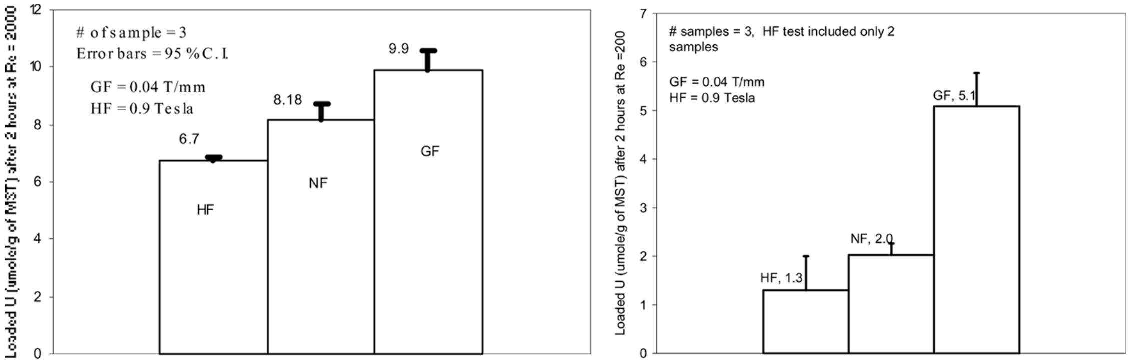

FIG. 4. This figure shows the effect of magnetic field geometry on Uranium loading at 300 rpm (left figure) and 30 rpm (right figure). The magnetic field gradient enhanced the sorption rate and the homogeneous magnetic fields did not (NF = no field, HF = homogeneous field, and GF = gradient field).

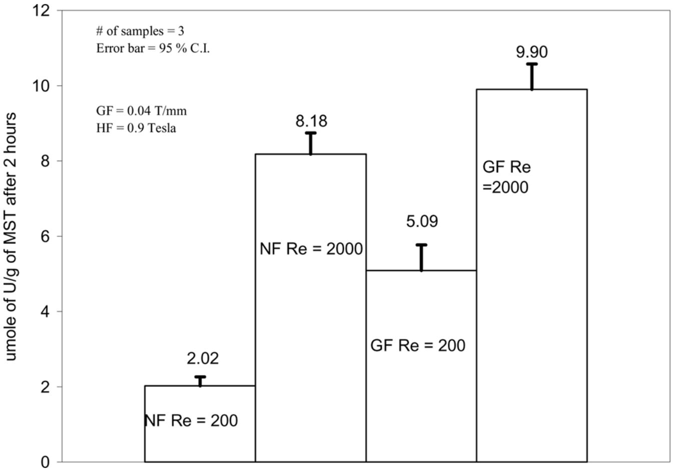

FIG. 5. The effect of mixing on uranium loading rate on MST (after 2 hours exposure to MST) shows that the magnetic field gradient enhanced sorption just as bulk mixing (NF = no field and GF = gradient field).

uranium (and strontium) adsorption. Also note that the data in Fig. 4 shows that higher mixing energy (higher spinning rate) did not improve sorption under a homogeneous magnetic field. The strontium data measured below detection limit at high Reynolds number (2000). This is due to the fact that $SrOH^+$ or Sr^{2+} diffuse faster than the anionic uranium.

Mixing effects on U sorption on MST is shown in Fig. 5. Visual inspection of Fig. 5 reveals that increasing the mixing rate (Reynolds number's from 200 to 2000) increased the U sorption on MST by as much as a factor of 2 under a gradient magnetic field and by as much as a factor of 4 under no magnetic field.

DISCUSSION

Researchers envision at least four mechanisms that can enhance the uranium sorption rate on MST under a gradient magnetic field.

These include:

1. additional convective bulk flow due to a electrically conductive salt solution moving in a magnetic field,
2. enhanced uranium diffusion due to a high gradient magnetic field around the MST particles,
3. a disruption of the hydrogen bonded water network (outside and inside the MST particles), and
4. enhanced sorption due to increased MST particle mobility due to the interaction of the magnetic field.

We do not believe the observed phenomena is due to an increase in the MST particle mobility because the relatively large size of the MST particles and the low magnetic susceptibility of the chemically similar TiO_2. The magnetic susceptibility of titanium dioxide is approximately 9.24 E-7 emu/g (8). The combination of the small magnetic susceptibility and a magnetic field of 0.9 Tesla does not generate enough force to move or lift the MST particles having a particle size of 1 to 30 microns diameter. Therefore, no additional convective flow is expected from the translation or rotation of the MST particles that may result from interactions with the magnetic field gradient. In addition, due to the small magnetic susceptibility, there is no magnetic field enhancement near the MST particles (i.e., concentration of the magnetic field lines as they traverse a MST particle) as seen in High Gradient Magnetic Separation tests (9). Therefore, researchers do not expect any sorption enhancement that may result from the magnetic susceptibility of MST.

Currently the available data do not allow us to attribute the observed phenomena to one or more of the remaining three phenomena mentioned above.

CONCLUSION

The presence of a magnetic field gradient enhanced the rate of sorption of strontium and uranium on MST. The enhancement was evident in the early contact times. No enhancement was seen when the sorption test was conducted under a homogeneous magnetic field.

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