Environ. Sci. Technol. **1998**, *32,* 2628−2632

Long-Term Chemical and Biological Stability of Surfactant-Modified Zeolite

ZHAOHUI LI,*,† STEPHEN J. ROY,† YIQIAO ZOU,‡ AND ROBERT S. BOWMAN†

Department of Earth and Environmental Science, Department of Chemistry, New Mexico Institute of Mining and Technology, Socorro, New Mexico 87801

We determined the chemical and biological stability of surfactant-modified zeolite (SMZ) in laboratory batch and column experiments. SMZ prepared using hexadecyltrimethylammonium (HDTMA)-Br or -Cl was stable in high ionic strength and high- and low-pH environments, with more than 90% of HDTMA remaining bound to the zeolite surface after washing with 100 pore volumes. However, up to 20% of the HDTMA washed off in low ionic strength solutions (I < 5 mM) and up to 50% washed off in a 1.0 mM Cs^+ solution. HDTMA bound to the zeolite surface was resistant to microbial degradation, with more than 98% of the original HDTMA remaining after 12−17 weeks of incubation under aerobic or anaerobic conditions. While aqueous HDTMA was biocidal, HDTMA bound on SMZ did not inhibit microbial growth. On the basis of the results of this study, SMZ appears suitable as a sorbent for long-term in situ applications and as a substrate for enhanced bioremediation.

Introduction

Clay minerals and zeolites have permanent negative charges on their surfaces, which enables them to be modified by cationic surfactants to enhance contaminant retention and retard contaminant migration (*1*−*5*). The advantages of using such materials are their low cost and high contaminant removal. While surfactant-modified clay minerals have been proposed as impermeable barriers for landfill liners, the excellent hydraulic properties of surfactant-modified zeolite (SMZ) make it an ideal candidate for water treatment in flow-through applications (*6*). Laboratory batch and column tests demonstrate that SMZ can simultaneously remove multiple types of contaminants from water, including inorganic anions such as chromate and hydrophobic organics such as chlorinated solvents and fuel components (*4*, *6*−*9*). Upon the basis of this earlier work, a pilot scale demonstration of an SMZ subsurface permeable barrier for groundwater remediation is currently in progress (*10*). The ultimate usefulness of surfactant-modified minerals depends not only upon their efficiency of contaminant removal from water but also upon their long-term chemical and biological stability.

The cationic surfactant hexadecyltrimethylammonium (HDTMA), one of a group of quaternary ammonium compounds (QACs), is commonly used for surface modification. It has been found that both the initial amount of surfactant in the system and the initial surfactant concentration affect the amount of surfactant sorbed (*8*, *11*). At very low concentrations below the critical micelle concentration (CMC), which is typically <1 mM, the surfactant molecules exist as monomers in aqueous solution. When the surfactant concentration is greater than the CMC, the surfactant molecules associate to form solution micelles in addition to monomers. If the initial surfactant concentration is greater than the CMC, the surfactant molecules sorbed on clay minerals or zeolite form bilayers, with the first layer retained by cation exchange and the second layer by hydrophobic bonding and stabilized by counterions (*11*, *12*). Increases in ionic strength promoted HDTMA sorption via hydrophobic bonding by lowering the CMC (*11*). Changes in counterions from more hydrated to less hydrated enhanced the bilayer stabilization (*12*). Therefore, the stability of sorbed surfactant on zeolite can be evaluated by studying desorption as a function of counterion species and ionic strength. After a period of 7 days shaking with 0.1 M KCl, desorption of HDTMA from montmorillonites was less than 1% when the treatment was below 150% of the cation-exchange capacity (CEC) and 9.4% when the treatment was 200% of the CEC (*3*). In a study of the cation exchange of HDTMA in a subsoil containing vermiculite, it was found that the most stable HDTMA−soil complexes formed at HDTMA loadings of 0.6−0.7 CEC for nonsodic soils (*13*). In a column study using Borden aquifer material treated to 33% of the CEC with HDTMA, all the HDTMA remained sorbed in the column after flushing with 325 pore volumes of surfactant-free water (*5*). All of the above observations suggest that HDTMA sorbed on clays and soils is stable with respect to desorption. Similarly, SMZ was shown to be stable in a range of different liquid media for at least 72 h (*6*).

The resistance of surfactant-modified surfaces to biological attack is also of importance. Although many QACs are biocidal (*14*), they can be degraded microbially. Tests on biodegradation of decyltrimethylammonium bromide indicated that it was utilized as a sole carbon source by a mixed population of two bacteria isolated from soil and wastewater (*15*). Ditallowdimethylammonium chloride and distearydimethylammonium chloride in aqueous solution were mineralized under aerobic conditions (*16*). At initial concentrations between 0.1 and 20 mg/L, extensive biodegradation of octadecyltrimethylammonium chloride in aqueous solution was noticed (*17*). Van Ginkel et al. (*18*) isolated a *Pseudomonas* sp. from activated sludge that utilizes HDTMA chloride as a carbon and energy source. Degradation of HDTMA in aqueous solution was complete in 70 h and resulted in stoichiometric formation of trimethylamine (*18*). On the other hand, only a limited amount of HDTMA-Br in aqueous solution was decomposed after 60 days by organisms when the initial concentration was 10 or 25 mg/L, and no decomposition was found when the initial concentration was 100 mg/L (*15*). In the studies cited above, biodegradation of QACs was observed only in solution. No studies of biodegradation of sorbed HDTMA have been reported.

Microbial toxicity studies of HDTMA in solution and sorbed to soil indicated that aqueous HDTMA-Br added to soil increased the lag periods and decreased the rate and extent of mineralization of test compounds. This result was interpreted as selective toxicity toward Gram-negative soil microorganisms (*19*). Relative to aqueous HDTMA, however, once bound to soil or clay, HDTMA toxicity was greatly reduced (*19*). Since HDTMA is approved for use as an additive in hair conditioner, mouthwash, and fabric softener (*6*), it is assumed that low levels of HDTMA will not be harmful to

* To whom correspondence should be addressed. E-mail: zli@nmt.edu; fax: (505)835-6436; phone: (505)835-5466.
† Department of Earth and Environmental Science.
‡ Department of Chemistry.

S0013-936X(97)00841-9 CCC: $15.00
Published on Web 07/30/1998

the environment. On the other hand, due to the biocidal properties of QACs, the effect of surfactant-modified materials on common microorganisms needs to be determined.

To date, no comprehensive studies of the chemical and biological stability of surfactant-modified minerals and soils have been reported. The objective of this research was therefore to determine the long-term chemical and biological stability of SMZ under aggressive and environmentally relevant conditions and to determine the impact of SMZ on environmental microorganisms.

Materials and Methods

SMZ Preparation. The zeolite used in this study is a clinoptilolite from the St. Cloud mine in Winston, NM. Detailed mineralogical characterization can be found elsewhere (*12*). HDTMA-Br solution (0.066 M) or 0.05 M HDTMA-Cl solution (both from Aldrich, Milwaukee, WI) was spiked with ^{14}C-methyl-labeled HDTMA-I with a specific activity of 110 mCi/mM (American Radiolabeled Chemicals Inc., St. Louis, MO) to yield a count rate of 17 000−22 000 cpm/mL. One-hundred seventy milliliters of solution and 67 g of raw zeolite were put into each of two 500 mL polyethylene centrifuge bottles and shaken for 8 h, which preliminary experiments showed sufficient for attaining sorption equilibrium. Experimental blanks showed that HDTMA did not sorb to plastic labware including polyethylene, polyallomer, and acrylic. The mixture was then centrifuged at 14 500*g* for 25 min followed by washing with two portions of 170 mL of type I water (18 MΩ deionized water from a Milli-Q system, Millipore Corp., Bedford, MA). For each washing, the bottles were shaken at 150 rpm for 15 min and centrifuged at 14 500*g* for 25 min. The samples were air-dried following the final washing. All the supernatants were analyzed for [^{14}C]HDTMA aqueous concentration by adding 1 mL of supernatant to 9 mL of scintillation cocktail (ICN Biomedicals, Inc., Costa Mesa, CA) and counting on a Packard Tri-Carb liquid scintillation counter. The amount of HDTMA sorbed was calculated from the difference between the initial and final concentrations. The HDTMA loading on the zeolite was 196 mequiv/kg for HDTMA-Br and 143 mequiv/kg for HDTMA-Cl, very close to the targeted loadings of 200 and 150 mmol/kg, which are the HDTMA sorption capacities for each counterion and are optimums for contaminant sorption by SMZ (*12*). The pH values of the final solutions were 7.0 ± 0.2 for HDTMA-Br and 6.6 ± 0.2 for HDTMA-Cl.

Chemical Stability of SMZ. The chemical reagents used for the stability experiments were 0.1 M $Na_2S_2O_4$ and 5% H_2O_2 solutions to simulate reducing and oxidizing conditions; 1.0 M CsCl and 1 M $CaCl_2$ solutions to represent high ionic strengths with monovalent and divalent cations; type I water to represent low ionic strength conditions; and pH 3 buffer (0.05 M potassium hydrogen phthalate and 0.02 M HCl) and pH 10 buffer (0.025 M $NaHCO_3$ and 0.01 M NaOH) to represent low and high pH conditions. Two and one-half grams of ^{14}C-labeled SMZ and 15 mL of each reagent were put into a 50 mL polyallomer centrifuge tube and shaken at 25 °C and 150 rpm. After 24 h, a time sufficient to attain desorption equilibrium (*20*), the mixture was centrifuged at 29 100*g* for 15 min to yield a clear supernatant. Ten milliliters of the supernatant was removed, and 1 mL was analyzed for equilibrium HDTMA concentration using liquid scintillation counting as described above. Then 10 mL of fresh reagent was added to the mixture and the mixture reshaken. The desorption cycle was repeated 11 times. Each experiment was performed in duplicate. Using the zeolite bulk density of 0.9 g/cm^3 and the porosity of 0.6 (which includes both primary porosity between zeolite aggregates and secondary porosity within the zeolite aggregates and individual zeolite crystals) as measured in laboratory transport experiments (*7*), each sample was washed by the equivalent of 110 pore volumes of reagent.

To simulate leaching by water of different ionic strengths, HDTMA desorption was also tested in flow-through column experiments. A series of minicolumn studies were conducted where 2.5 g of SMZ (150 mmol/kg HDTMA-Cl) was put into a 5 mL polyethylene column (0.8 cm internal diameter and 5 cm in length) and leached with aqueous $NaHCO_3$ solutions having concentrations of 1, 5, 10, or 15 mM. The equivalent of 90 pore volumes of solution was added to each column in 30 equal-volume slugs. Each slug of fluid was allowed to drain by gravity prior to the addition of the next slug. Longer-term leaching studies under steady flow conditions were performed using 30 cm long, 5 cm diameter acrylic columns packed with SMZ. The leaching solution was 0.5 mM $NaHCO_3$ with a specific discharge of 19.4 cm/day (mean pore water velocity of 32.3 cm/day). The hydraulic conductivity of the SMZ was 10^{-3} m/s (*7*). Samples were collected on 2−7 day intervals for approximately 5 months and analyzed for HDTMA by an HPLC method (*12*). All column studies were done in duplicate.

Biological Stability of SMZ. The experiments were performed in modified Bartha-Pramer flasks (*21*) to determine the mineralization of ^{14}C-labeled HDTMA by microorganisms. Microcosms were used under operationally defined aerobic, anaerobic, saturated, and unsaturated conditions for HDTMA-Cl and HDTMA-Br treated zeolite as well as aqueous solutions of HDTMA. The microcosms consisted of 50 mL Erlenmeyer flasks with a hanging cup for storage of the alkaline solution used to trap $^{14}CO_2$. For the unsaturated aerobic condition, 5 g of [^{14}C]SMZ, 2 mL of nutrient solution [2 g of K_2HPO_4, 7 g of KH_2PO_4, 1 g of $(NH_4)_2SO_4$, 0.1 g of $MgSO_4 \cdot 7H_2O$, 0.01 g of $FeSO_4 \cdot 5H_2O$/1000 mL of type I water], and 2 mL of activated sludge (from the Socorro, NM sewage treatment plant) were put into the flask. The water content was 70% of saturation. For the saturated aerobic condition, 5 g of [^{14}C]SMZ, 5 mL of nutrient solution and 5 mL of activated sludge were used. In this treatment, the SMZ was totally submerged in the solution. The saturated anaerobic treatment was the same as the saturated aerobic treatment except that the headspace was flushed with nitrogen gas each time the system was opened for sampling. For the HDTMA aqueous solution treatment, 0.1 mL of ^{14}C-labeled HDTMA-I was added to 50 mL of 0.66 mM HDTMA-Br or 50 mL of 0.5 mM HDTMA-Cl solution to yield an activity of 0.011 μCi/mL. These aqueous microcosms consisted of 5 mL of HDTMA solution, 2 mL of activated sludge, and 2 mL of nutrient solution. All the microcosms were incubated at room temperature (22−23 °C) in the dark. The alkaline solution in the hanging cup (0.3 mL of 0.3 M NaOH) was removed and replaced with a fresh solution every 2 weeks. The alkaline solution was immediately put into 10 mL scintillation vials containing 9 mL scintillation cocktail and analyzed for $^{14}CO_2$ released due to the mineralization of HDTMA. Each experimental condition was replicated four times. Poisoned controls (10 mM $HgCl_2$) were included for the same initial conditions. The biological stability experiments lasted 12 weeks for the unsaturated conditions and the HDTMA aqueous solution and 17 weeks for the saturated conditions.

Biological Toxicity of SMZ. Every 2 weeks during the biological stability experiment, agar plates were inoculated with the mixture from each microcosm to check the viability of the microorganisms. The inoculated plates were incubated at room temperature for 72 h in the dark. At the end of the incubation, microorganism growth was recorded according to the number of the colonies on the plates and the percentage of the agar plates covered by biomass.

Chromate Sorption to SMZ. The original SMZ and the SMZ recovered from each long-term biological stability study were tested for chromate sorption. Detailed experimental

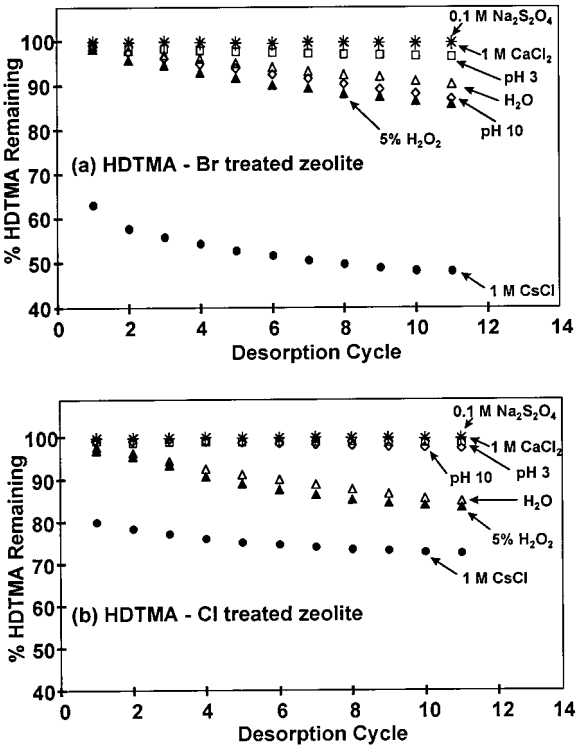

FIGURE 1. HDTMA remaining on the zeolite surface after 11 desorption cycles with different reagents. Symbols are averages of duplicates.

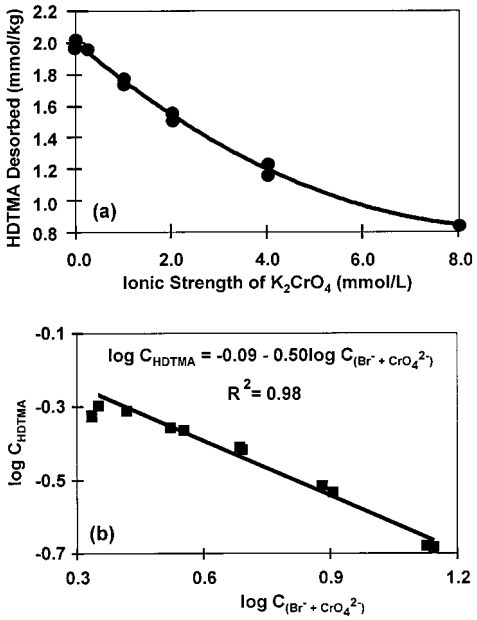

FIGURE 2. Effect of ionic strength on HDTMA desorption from zeolite. (a) Amount desorbed by 6 pore volumes of aqueous K_2CrO_4. (b) Desorbed HDTMA concentration vs total anion concentration (Br^- + CrO_4^{2-}) in solution.

and analytical methods can be found elsewhere (*12*). All sorption experiments were performed in duplicate. Sorbed chromate was calculated from the difference between initial and final solution concentrations.

Results and Discussion

The results of long-term chemical stability of SMZ after 11 desorption cycles with different reagents can be seen in Figure 1. For each washing solution at each time, replicates varied by less than 5% from the mean value. The desorption of HDTMA follows CsCl > H_2O_2 > H_2O > pH 10, pH 3, $CaCl_2$ > $Na_2S_2O_4$. CsCl removed 20−35% of the sorbed HDTMA during the first desorption cycle. At the end of the experiment, 50% of the initial sorbed HDTMA had desorbed when Br^- was the counterion (Figure 1a) while 30% desorbed when the counterion was Cl^- (Figure 1b). With an initial HDTMA loading of 196 mmol/kg and 143 mmol/kg for HDTMA-Br and HDTMA-Cl modified zeolites, the remaining HDTMA after 1 M CsCl desorption is 100 mmol/kg and 104 mmol/kg, respectively, which corresponds to a monolayer coverage of HDTMA regardless of the type of counterion. While HDTMA desorption by 1.0 M Cs^+ is significant, in practice, such high concentrations of Cs^+ are rarely encountered in even highly contaminated systems.

Desorption of HDTMA by H_2O (10−15%) was probably due to the low ionic strength of the system which caused the electric double layer to expand and weaken the interaction between the HDTMA headgroups and the counterions. The effect of ionic strength on HDTMA desorption at fixed solution volume (6 pore volumes) was tested using K_2CrO_4 as the ionic strength adjuster. Zeolite prepared with HDTMA-Br was tested. HDTMA desorption was 0.34 mmol/kg per pore volume when type I water ($I = 0$) was used and reduced to 0.14 mmol/kg/pore volume when aqueous K_2CrO_4 concentration was 2.8 mmol/L or $I = 8$ mM (Figure 2a). On the basis of extrapolation of the curve in Figure 2a, after washing with 500 pore volumes of a solution at the ionic strength of 8 mM, the HDTMA desorption would be ~70 mmol/kg, and 65% of sorbed HDTMA would still remain on the surface. The prediction was verified in a laboratory column test for chromate retardation by SMZ, in which 65% of the initial sorbed HDTMA remained on the surface after flushing with about 500 pore volumes of water at an ionic strength of 9 mM (*7*).

The influence of total anion concentration (C_{anion}) on the CMC of cationic surfactants can be described by (*22*)

$$\log \text{CMC} = a' + b' \log C_{\text{anion}} \quad (1)$$

where a and b ($b < 0$) are constants. Xu and Boyd (*11*) indicated a similar relationship to predict the residual HDTMA concentration (a_{HDTMA}) during HDTMA sorption on vermiculitic subsoil

$$\log a_{\text{HDTMA}} = a + b \log a_{\text{anion}} \quad (2)$$

in which a and b are constants and a_{anion} is the anion activity. A plot of the logarithm of HDTMA aqueous concentration after 24 h desorption vs the final anion concentration as determined by Br^- and CrO_4^{2-} analysis can be seen in Figure 2b. The intercept (−0.09) corresponds to an HDTMA concentration of 0.8 mM, very close to the CMC of HDTMA-Br in pure water (0.9 mM). The similarity between eqs 1 and 2 may indicate that both equations reflect the same relationship between surfactant concentration and the counterion activity. The relationships suggest that only monomers desorb from the sorbed bilayer and the amount of monomer desorbed depends on the anion concentration in solution. This would explain why more HDTMA desorbed when SMZ was equilibrated with water than with higher ionic strength solutions.

Column leaching experiments for both the minicolumns and 30 cm columns show that after flushing with 90 pore volumes, 25−35 mmol/kg HDTMA desorbed, which translates into 100−110 mmol/kg of HDTMA (or 65−75%) still remaining on the surface (Figure 3). The amount of HDTMA remaining on the surface is again in rough agreement with monolayer coverage (100 mmol/kg). Since the monolayer or the lower layer of the bilayer is held primarily by

TABLE 1. Microorganism Growth on Agar Plates after Inoculation with the Mixture of SMZ and Activated Sludge[a]

experimental conditions	week 2	week 4	week 12	week 17
unsaturated, aerobic, HDTMA-Cl-treated SMZ	+, p, g, s, l	++, p, g, s, l	++, p, g, s, l	++, p, g, s, l
unsaturated, aerobic, HDTMA-Br-treated SMZ	+, p, g, s, l	++, p, g, s, l	++, g, s, l	++, p, s, l
unsaturated, aerobic, HDTMA-Cl-treated SMZ with Hg poisoning	+, p, s, l	+, p, g, s, l	++, p, g, s, l	++, g, s, l
unsaturated, aerobic, HDTMA-Br-treated SMZ with Hg poisoning	+, p, s, l	++, p, g, s, l	++, p, g, s, l	+++, g, s, l
aqueous 0.5 mM HDTMA-Cl solution	−	−	−	−
aqueous 0.66 mM HDTMA-Br solution	−	−	−	+, g, l
aqueous 0.5 mM HDTMA-Cl solution with Hg poisoning	−	−	−	−
aqueous 0.66 mM HDTMA-Br solution with Hg poisoning	−	−	−	−
saturated, aerobic, HDTMA-Cl-treated SMZ	+, g, s, l		++, g, s, l	
saturated, aerobic, HDTMA-Br-treated SMZ	+, g, s, l		++, g, s, l	
saturated, anaerobic, HDTMA-Cl-treated SMZ	+, p, g, s, l		++, g, s, l	
saturated, anaerobic, HDTMA-Br-treated SMZ	+, g, s, l		++, g, s, l	

[a] (−) No bacteria growth; (+) bacteria growth; (++) enhanced bacteria growth; (+++) greatly enhanced bacteria growth; (p) pink colony; (g) gray colony; (s) small colony; (l) large colony.

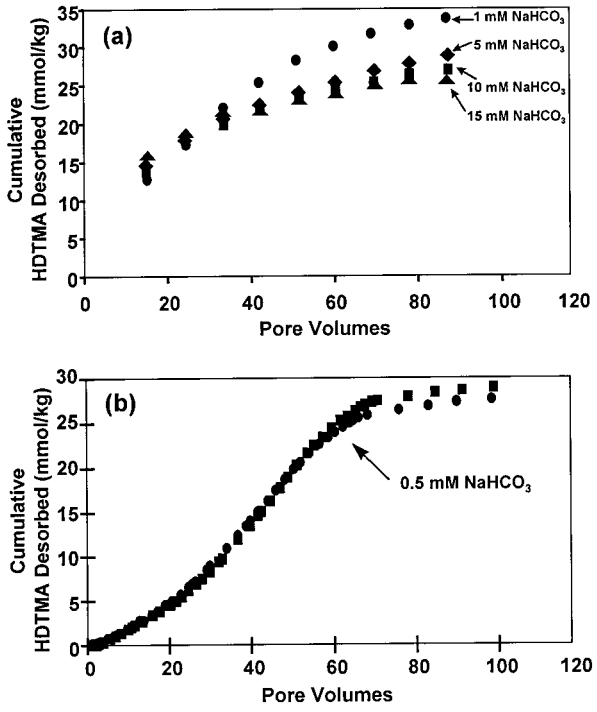

FIGURE 3. HDTMA desorption vs pore volume. (a) Leaching through minicolumns with different aqueous $NaHCO_3$ concentrations. Symbols are averages of duplicates. (b) Leaching through duplicate 30 cm columns with 0.5 mM $NaHCO_3$ over 5 months.

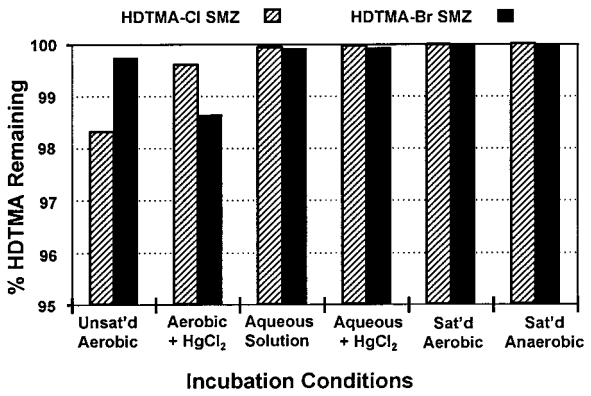

FIGURE 4. HDTMA remaining on SMZ after 12−17 weeks of incubation under various conditions. Each bar is the average of four replicates.

electrostatic forces and the upper layer of the bilayer is held primarily by hydrophobic bonding and stabilized by counterions (*11*, *12*), it may be inferred that the HDTMA−zeolite interaction for monolayer surface coverage is much stronger than for bilayer coverage. Brown and Burris (*5*) noted similar results while working with surfactant-modified Borden sand treated to below-monolayer coverage. The results in Figure 3a also indicate that higher ionic strength caused less HDTMA desorption, in agreement with the batch stability results.

Desorption of HDTMA from SMZ in aqueous solutions is comparable to that from clays. At an HDTMA loading of 1.95 times the CEC of the vermiculitic subsoil, Xu and Boyd (*11*) found that the fraction of HDTMA desorbed in 13 washes varied from 8 to 29% and was strongly dependent on ionic strength, with the greatest desorption occurring in pure H_2O. The solid-to-liquid ratio they used was 1:10 (w/w), which would give 20 pore volumes per wash assuming the porosity of the material was 0.5 and the bulk density was 1.0 g/cm^3. After washing with 100 pore volumes of water, the fractional HDTMA remaining on the surface was 0.83 (initial HDTMA loading = 1.95 CEC), which is comparable to the results of our research (81−85% in Figure 1a).

Since the desorption by H_2O_2 was almost the same as by water (15−17%), the desorption may also be due to the low ionic strength in the system. Under reducing conditions as simulated by washing with $Na_2S_2O_4$, there was essentially no removal of sorbed HDTMA. Nor was HDTMA removed by 1.0 M $CaCl_2$, although Ca^{2+} is a divalent cation. Similar results were observed in a desorption study of HDTMA-modified montmorillonite in 0.1 M NaCl or KCl solutions (*3*). Under acidic conditions, the desorption of HDTMA was negligible (Figure 1). Under basic conditions, the desorption was negligible when Cl^- was the HDTMA counterion, while the desorption was 15% when Br^- was the HDTMA counterion. Another striking characteristic is that more desorption occurred when Br^- was the counterion than when Cl^- was the counterion, except for the low ionic strength cases. This may also indicate greater monolayer stability than bilayer stability since HDTMA loading was 33% higher when Br^- was the counterion.

The results of the biological stability experiments are plotted in Figure 4. After 12−17 weeks incubation with activated sludge, >98% of the initial HDTMA remained on the surface of the SMZ in all treatments. The poison control ($HgCl_2$) was not effective, due to the sorption of mercury from solution by SMZ, which was verified in a separate mercury sorption experiment (Li and Bowman, unpublished data).

The results of the SMZ toxicity experiments indicated that the bacteria remained viable in all the microcosms with SMZ. In those microcosms containing aqueous HDTMA without zeolite, HDTMA inhibited the growth of the microorganisms (Table 1). The results were in agreement with the toxicity

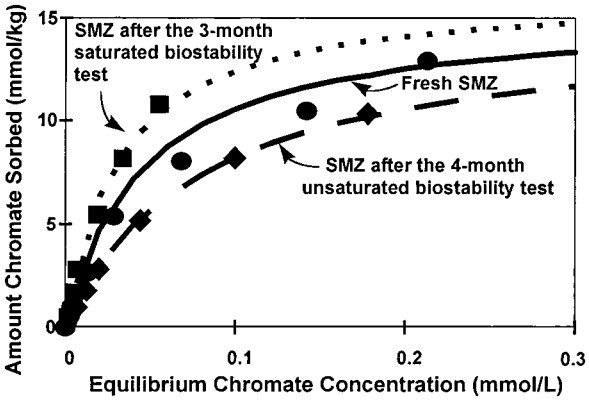

FIGURE 5. Chromate sorption on HDTMA-Cl SMZ before and after the long-term biological stability tests. The lines are Langmuir fits to the observed data. Symbols are averages of duplicates.

experiment using HDTMA-smectite as observed by Nye et al. (*19*).

At the end of the biological stability experiments, the SMZ was dried and evaluated for chromate sorption. The assumption was that if HDTMA was biodegraded but no $^{14}CO_2$ was evolved, the sorption of chromate would be greatly reduced. A comparison of chromate sorption on SMZ before and after the biostability experiment can be seen in Figure 5. As described by Haggerty and Bowman (*4*), chromate sorption by SMZ follows a Langmuir sorption isotherm. The chromate sorption capacity of SMZ (from Langmuir fits to the data of Figure 5) after exposure to microorganisms for 12−17 weeks remained essentially unchanged; this further confirms that sorbed HDTMA is biologically stable, since there would have been no chromate sorption had the sorbed HDTMA bilayer been degraded by microorganisms.

The results of this study demonstrate that SMZ exhibits long-term chemical and biological stability over a wide range of pH, Eh, aerobic, anaerobic, saturated, and unsaturated conditions. SMZ does not appear to affect microbial activity and thus should have minimal impacts when used for in situ applications such as permeable barriers. Additionally, the lack of microbial toxicity may suggest that SMZ could be a useful substrate for enhanced bioremediation.

Acknowledgments

This research was supported by the U.S. DOE under Contract DE-AR21-95-MC32108 from the Federal Energy Technology Center. Todd Burt and Kirk Jones of New Mexico Tech performed some of the desorption and biostability experiments.

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Received for review September 19, 1997. Revised manuscript received June 15, 1998. Accepted June 25, 1998.

ES970841E