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Particle Release and Permeability Reduction in a Natural Zeolite (Clinoptilolite) and Sand Porous Medium

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To evaluate clinoptilolite, a natural zeolite, as a candidate material for a permeable reactive barrier for removal of strontium from groundwater, we investigated particle release and permeability reduction in clinoptilolite and sand porous media. In flow-through column experiments, we tested the effects of solution chemistry, grain size, and clinoptilolite pretreatment on particle release and hydraulic conductivity. Permeability reduction occurred not in the clinoptilolite itself but only in finer-grained sand down-gradient of the clinoptilolite. Solutions of high ionic strength inhibited particle release and prevented clogging. Clinoptilolite of larger grain size produced slightly less particle release and clogging. Two pretreatments of the clinoptilolite, rinsing to remove fine particles and calcining to improve strength, reduced particle release and clogging. Calcining, however, significantly reduced the strontium binding strength of the clinoptilolite.

Introduction

Over the last few decades, several spent nuclear fuel storage sites in the United States have suffered significant groundwater contamination by ^{90}Sr. This radioactive cation is abundant in spent nuclear fuel and relatively mobile in sand and gravel aquifers with low clay content. To prevent further migration of ^{90}Sr, the installation of a permeable reactive barrier to remove strontium from contaminated groundwater has been considered at the Hanford Nuclear Reservation (*1*) and implemented on a pilot scale at the West Valley Demonstration Project (*2*). For a reactive barrier to be successful, strontium must be adsorbed in the presence of competitive cations and retained in the barrier for many half-lives (^{90}Sr, 28.5 y). In addition, barrier permeability must be maintained so contaminated groundwater will freely flow through, not around, the barrier (*3*, *4*).

The first criterion, to adsorb radioactive cations, is met using zeolites as the barrier material. Zeolites have been used in ex situ ion exchange beds to remove strontium, cesium, ammonium, and heavy metals from wastewaters, even in the presence of more abundant competing cations (*5*−*11*). Clinoptilolite, the most cost-effective zeolite for removal of radioactive cations from wastewaters (*12*), strongly adsorbed strontium in West Valley, NY, and Hanford, WA, groundwaters (*1*, *13*) and was selected as the material for the West Valley pilot barrier (*2*).

The second criterion, to retain the radioactive cations for many half-lives, may not be so readily met by zeolites. In behavior that has been attributed to their brittle nature, zeolites release fine particles and with them, presumably, adsorbed cations. Various zeolites suffered significant weight loss, or “attrition” (1−18%), during backwash and regeneration in ion exchange beds (*6*, *14*). Klieve and Semmens (*15*) suggested that the extent of attrition was related to the crushing resistance of the zeolite. Weaker zeolites such as phillipsite would be expected to show greater attrition; stronger zeolites such as clinoptilolite would show less attrition.

Particle release in a zeolite barrier may lead to a more serious problem—clogging. In ion exchange beds, zeolite attrition caused head loss that could be remedied only by hydraulic lifting or media replacement (*7*). If particle release leads to permeability reduction in a reactive barrier, the contaminated groundwater would be diverted around the barrier. In various porous media, particle release caused by decreases in ionic strength during artificial recharge (*16*), secondary oil recovery (*17*), and freshwater-seawater mixing (*18*) have been implicated in clogging. The extent of clogging has been related to the abundance of fine particles in the aquifer material (*18*, *19*).

In this study, we evaluated the effects of ionic strength, grain size, and two pretreatments, rinsing and calcining (heating), on particle release and clogging in a clinoptilolite porous media. We expected rinsing to minimize particle release by removing fine particles (*6*). Calcining (*15*) was expected to reduce particle release by improving strength through dehydration (*20*). In addition, we measured the strontium binding strength of the untreated and treated clinoptilolites to determine if the pretreatments would reduce the ability of the clinoptilolite to adsorb cations.

Materials and Methods

Natural Zeolite. Clinoptilolite (type CH, Teague Mineral Products, Adrian, OR) was obtained in two size fractions, fine (−20/+50 mesh; 0.30 to 0.85 mm) and coarse (−5/+20 mesh; 0.85−4.0 mm). The same clinoptilolite (−14/+50 mesh) was used in the West Valley Demonstration Project pilot barrier (*2*). Two pretreatments, rinsing and calcining, were applied to both size fractions. For rinsing, clinoptilolite (300 g) was suspended in high-purity water (600 mL; >18 Mohm resistivity) and allowed to settle for 60 s. The turbid supernatant was decanted and replaced with more water; this procedure was repeated about 20 times to produce a clear supernatant (turbidity <1.0 NTU). For calcining, clinoptilolite was heated at 550 °C for 48 h. Each of the pretreated clinoptilolites was equilibrated with 1×10^{-2} M $CaCl_2$ (pH 5.6 ± 0.1) solution for 72 h.

Sand. The sand was obtained from Unimin Corp. (Emmett, ID) in two different size ranges, fine (30% retained on 70 mesh; $d_{10} = 0.12$ mm) and coarse (60% retained on 40 mesh; $d_{10} = 0.37$ mm). The well-sorted angular to subangular sands contain about 80% quartz, 11% feldspars, 8% muscovite/biotite, and 1% other granitic lithoclasts.

Column Packing. Particle release and clogging were measured in columns packed with (a) clinoptilolite only and (b) separate layers of clinoptilolite and sand (Figure 1). Separate layers of sand and clinoptilolite were poured into the column partially filled with 1×10^{-2} M $CaCl_2$ solution in 2 cm lifts, while the column was lightly tapped. A nylon mesh (0.15 mm opening) placed on the influent end cap secured grains in the column. In both column arrangements, the overall porosity was 0.43−0.46.

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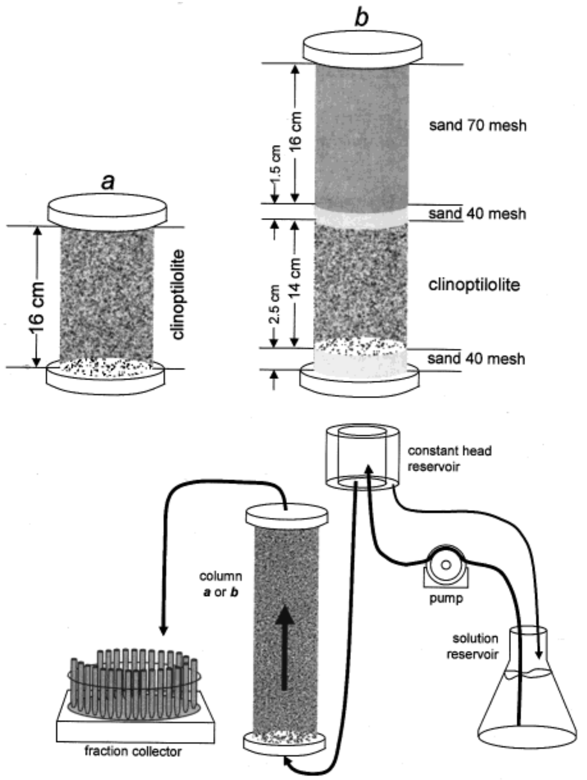

FIGURE 1. Experimental setup showing the two column packing arrangements: (a) clinoptilolite-only and (b) clinoptilolite/sand.

Column Setup. Upward flow through the glass column (4.8 cm diameter, 34 cm length) was fed by a constant head reservoir (Figure 1). The constant head difference was adjusted to give an initial flow rate of 0.40 ± 0.01 mL min^{-1} (pore velocity 0.7 ± 0.1 m d^{-1}). Each column was prepared by flushing for 7 d with a 1×10^{-2} M $CaCl_2$ (pH 5.6) solution. During the 7 d column preparation, particle release was negligible (<1 mg L^{-1}), flow remained within 2% of the initial rate, and the effluent pH did not vary by more 0.3 pH units from the influent pH. The experiments commenced by changing the influent solution to 1.0×10^{-5} M NaCl (pH 5.6 ± 0.1) solution and continued until complete clogging or 21.1 (clinoptilolite-only columns) or 8.8 (clinoptilolite/sand columns) pore volumes of NaCl solution had been passed through the column.

Column Effluent Analysis. The mass of each effluent sample was measured to determine flow rate. The hydraulic conductivity was determined for each sample time period using the Darcy equation. The effluent turbidity was measured in a turbidity meter. Turbidity was converted to particle concentration using a linear regression determined by weighing particle mass after evaporation of water in 10 samples of turbidity ranging from 15 to 1890 NTU. The amount of particles released from the columns ("attrition") was calculated as a ratio of the total mass of particles released to the total mass of clinoptilolite in the column for each experiment.

Clinoptilolite Characterization. Scanning electron microscopy was used to examine gold-coated clinoptilolite samples mounted on aluminum stubs. The mineralogy of untreated and treated clinoptilolites and particles released from the clinoptilolite-only columns and trapped on 0.1 μm polycarbonate filters was determined by X-ray diffraction (XRD). The electrophoretic mobilities of clinoptilolite particles were measured by microelectrophoresis over a pH range of 2 to 11 in 1×10^{-2} M $CaCl_2$ and 1×10^{-5} M NaCl solutions. The size distribution (>1 μm) of particles released from clinoptilolite-only columns were measured by single-particle optical sensing. The crushing resistance of coarse clinoptilolite grains was measured by placing individual grains of 2–3 mm diameter between a balance and screw-driven plate and compressing the grains until failure was observed. Fifty grains were crushed; the five highest and lowest crushing forces were discarded, and the mean and standard deviation of 40 grains was reported as the crushing resistance.

Strontium Exchange. Strontium adsorption by calcium-saturated clinoptilolite was measured in batch experiments. Triplicate samples of clinoptilolite (1–10 g) in 1×10^{-3} M $SrCl_2$ solution (50 mL; pH 5.6) were prepared in polypropylene tubes, agitated on a rocking table for 72 h, and centrifuged at 14 000 g for 60 min. Supernatant was filtered through a 0.1 μm polycarbonate membrane, acidified with concentrated HNO_3 (1 mL), and analyzed for strontium on an inductively coupled plasma atomic emission spectrophotometer.

Results

Columns Containing Only Clinoptilolite. Particle release occurred in the clinoptilolite-only columns, but hydraulic conductivity did not decrease. The 1×10^{-2} M $CaCl_2$ solution, used for column preparation, caused very little particle release (maximum particle concentration of about 1.0 mg L^{-1}), while the 1×10^{-5} M NaCl solution caused substantial particle release (Table 1 and Figures 2 and 3). Rinsing and calcining decreased the amount of particles released. Slightly more particle release occurred in the fine clinoptilolite than in the coarse clinoptilolite.

Columns Containing Clinoptilolite and Sand. Both particle release and clogging occurred in the clinoptilolite/sand columns. Clogging occurred in the sand, not in the clinoptilolite. The amount of particles in the effluents of the clinoptilolite and sand columns were far less than those in the clinoptilolite-only column effluents owing to particle retention in the sand (Table 1). The trends in the particle release were similar to those observed in the clinoptilolite-only columns (Figure 4).

Clinoptilolite Characterization. The untreated and calcined clinoptilolite have similar distributions of fine particles on the grain surfaces, while rinsing removed fine particles (Figure 5). XRD revealed distinct peaks only for clinoptilolite in both the grains and the released particles, despite the smectite content reported by the supplier. Clinoptilolite mineralogy was not altered by either the rinsing or calcining treatment. At pH 6.0, clinoptilolite particles were more negatively charged in the 1×10^{-5} M NaCl solution (-1.5×10^{-6} μm s^{-1}/V cm^{-1}) than in the 1×10^{-2} M $CaCl_2$ solution (-0.2×10^{-6} μm s^{-1}/V cm^{-1}). The point of zero charge of the clinoptilolite particles was about pH 2. The size of the particles released from the columns containing only clinoptilolite roughly followed an inverse power law distribution over a range of 1 to about 50 μm (Figure 6). The crushing resistance of coarse clinoptilolite grains was improved slightly by calcining (untreated grains, 38 ± 3 N; calcined grains, 45 ± 4 N).

Strontium Exchange. The untreated fine clinoptilolite was most effective in exchanging Sr^{2+} (Table 2). More strontium was bound by the fine clinoptilolite than by the coarse clinoptilolite. Rinsing decreased the strontium distribution coefficient to about 55% of the untreated value. Calcining rendered the clinoptilolite much less effective at binding strontium.

Discussion

Mechanism of Particle Release. Low ionic strength caused extensive particle release, while high ionic strength caused little particle release (Figure 2). The effect of ionic strength on particle release highlights an important point—that

TABLE 1. List of Conditions and Results for Experiments Testing Particle Release and Permeability Reduction in Clinoptilolite-Only and Clinoptilolite/Sand Columns

column packing	clinoptilolite pretreatment	clinoptilolite grain size	no. reps[a]	K_{max} (cm s^{-1})	K_{min}/K_{max}[b]	pore volumes[c] at K_{min}/K_{max}	total particle release[d] (%)
clinoptilolite-only	untreated	fine	2	0.012 ± 0.001	0.99 ± 0.01	(21.1)	4.3 ± 0.2
	rinsed	fine	2	0.013 ± 0.001	0.98 ± 0.01	(21.1)	2.2 ± 0.3
	calcined	fine	1	0.012	1.0	(21.1)	0.060
	untreated	coarse	2	0.020 ± 0.001	0.99 ± 0.01	(21.1)	2.7 ± 0.1
	rinsed	coarse	2	0.020 ± 0.001	0.99 ± 0.01	(21.1)	1.5 ± 0.1
	calcined	coarse	1	0.021	1.0	(21.1)	0.057
clinoptilolite/sand	untreated	fine	3	0.0084 ± 0.0002	0.0 ± 0.0	4.3 ± 0.5	0.41 ± 0.10
	rinsed	fine	3	0.0083 ± 0.002	0.42 ± 0.13	(6.4 ± 0.4)	0.31 ± 0.06
	calcined	fine	3	0.0084 ± 0.002	0.99 ± 0.01	(8.8)	0.065 ± 0.012
	untreated	coarse	3	0.0085 ± 0.0003	0.0 ± 0.0	5.4 ± 0.2	0.25 ± 0.04
	rinsed	coarse	3	0.0085 ± 0.0002	0.60 ± 0.03	(5.2 ± 0.5)	0.21 ± 0.02
	calcined	coarse	3	0.0083 ± 0.0002	0.98 ± 0.01	(8.8)	0.039 ± 0.008

[a] Number of replicate experiments; replicates were conducted with fresh porous media. [b] The relative change in hydraulic conductivity recorded as the ratio of the minimum (K_{min}) and maximum (K_{max}) hydraulic conductivities, K_{min}/K_{max}, mean and one standard deviation. [c] The number of pore volumes at which K_{min}/K_{max} was recorded. Values in parentheses show number of pore volumes at which the experiment was completed. [d] Total particle release expressed as a percentage of the total mass of clinoptilolite in the column.

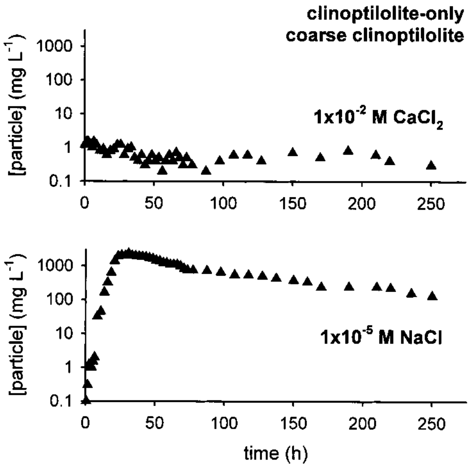

FIGURE 2. Effect of ionic strength and solution composition on particle release as a function of time in a clinoptilolite-only column containing coarse clinoptilolite. The first 250 h of the calcium chloride solution column pretreatment are shown.

electrostatic repulsion, not flow-induced shear forces, enables particle release. Low ionic strength enhances electrostatic repulsion and particle release (*21*−*24*). The lack of particle release by the high ionic strength solution indicates that shear is not the main cause of particle release. At the low flow velocity in the columns (0.69 m d^{-1}), shear was not expected to contribute to particle release. Shear on attached particles dominates release only at much higher flow velocities (*25*, *26*).

The ionic strength groundwater at the West Valley Demonstration Plant, the site of the pilot clinoptilolite barrier (*2*), is about 9.9×10^{-3} M. The major ions are Ca^{2+}, Na^+, Cl^-, and HCO_3^-, and the pH is near-neutral (*27*). While the sodium in the West Valley groundwater would favor particle release relative to calcium (*24*), this groundwater is sufficiently similar to the 1×10^{-2} M calcium chloride solution used in the column experiments to surmise that substantial particle release is not occurring in the pilot clinoptilolite barrier.

Mechanism of Permeability Reduction. Release of particles from the clinoptilolite caused reductions in hydraulic conductivity, or clogging, only when the fine-grained sand was present in the columns; therefore, clogging must have occurred in the sand. Reductions in hydraulic conductivity are typically caused by deposition of particles on grains (*28*), straining in pores (*29*, *30*), and swelling of smectite clay minerals such as montmorillonite (*18*, *31*). According to the supplier, smectites were present in the clinoptilolite, but we did not detect them in either the grains or the released particles; therefore, we assume that the released particles were predominantly clinoptilolite. Zeolites do not swell, so clogging in the sand must have been caused mainly by deposition or straining of the clinoptilolite particles. The sand is composed mainly of quartz and feldspar and both quartz and feldspar would be negatively charged at the pH of these experiments (*32*). Given the electrostatic repulsion that would exist between the clinoptilolite particles and sand surfaces, it unlikely that the particles were deposited on the sand grains at the low ionic strength of the solution in the column. Therefore, clogging must have been caused mainly by the straining.

The importance of straining as a particle removal method can be evaluated by comparing the size of the released particles to the size of the grains in the porous medium (*33*−*35*). If the grain size to particle size ratio, d_g/d_p, is less than 10, particles will not penetrate the porous medium and an impermeable layer forms on the porous medium surface. Such layers were not observed at the clinoptilolite/sand interface in the columns. For $10 < d_g/d_p < 20$, particles will enter the porous medium and be trapped in pores smaller than the particles. Sakthivadivel (*33*) observed order of magnitude reductions in hydraulic conductivity under these conditions. For $d_g/d_p > 20$, only slight reductions in hydraulic conductivity were observed.

For each porous medium, a critical particle size for straining can be identified with $d_g/d_p = 20$. For the coarse clinoptilolite, the minimum grain size is 0.85 mm, determined by sieving. The corresponding critical particle size is 43 μm (i.e., particles larger than 43 μm will be strained and cause clogging). For the fine clinoptilolite, the minimum grain size is 0.30 mm, and the critical particle size is 15 μm. The number of released particles of size greater than 15 μm is quite small (Figure 6), so it is unlikely that straining would occur in either the coarse or fine clinoptilolite.

The size distributions of the fine and coarse sand were not determined by sieving, so a minimum grain size will be approximated by the d_{10} values for these sands. For the coarse sand, the d_{10} value is 0.37 mm. The abundance of particles in the size range above the corresponding critical particle size, 19 μm, is small and clogging is not likely. The d_{10} value of the fine sand, 0.12 mm, results in a lowest critical particle size, 6 μm. At this critical particle size, particle abundance is about an order of magnitude greater than at the next lowest critical particle size, 15 μm, for fine clinoptilolite (Figure 6). The increase in abundance of particles that can be strained

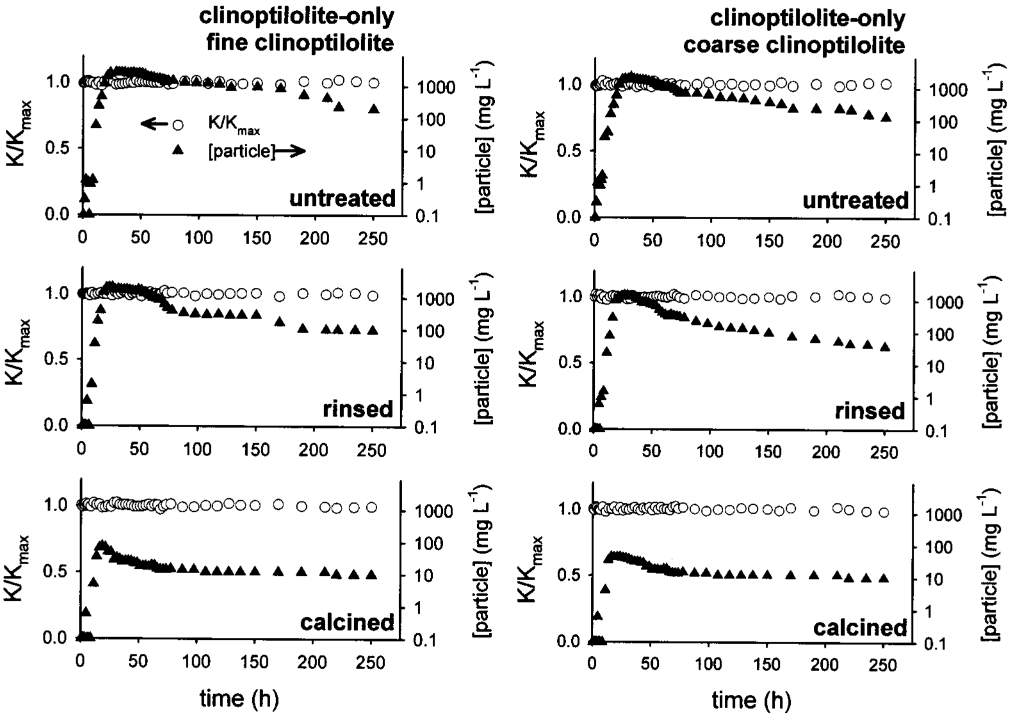

FIGURE 3. Effect of clinoptilolite pretreatment on the change in hydraulic conductivity (K/K_{max}) and particle release as a function of time in clinoptilolite-only columns filled with fine and coarse clinoptilolite. Results of one of duplicate columns for untreated and rinsed clinoptilolite; only experiment for calcined clinoptilolite.

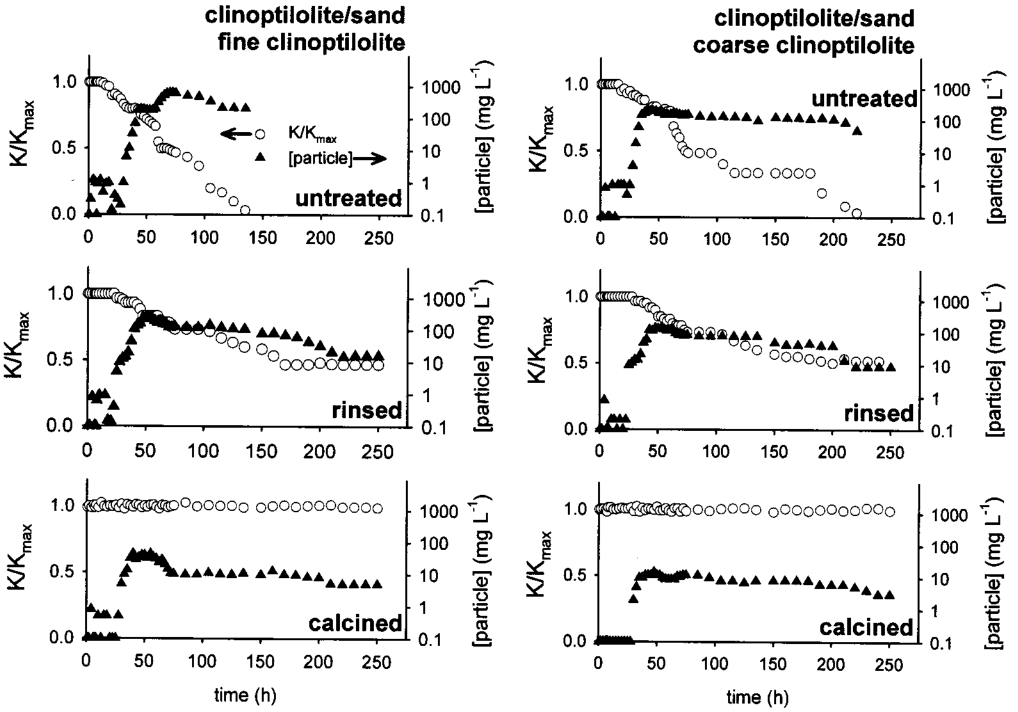

FIGURE 4. Effect of clinoptilolite pretreatment on the change in hydraulic conductivity (K/K_{max}) and particle release as a function of time in clinoptilolite/sand columns filled with fine and coarse clinoptilolite. Results of one of triplicate columns.

appears to be the cause of the clogging in the sand layers. It is important to remember that the d_g/d_p relationship was determined for spherical, uniform grains—both the clinoptilolite and sands are angular to subangular in shape and, at best, well-sorted, but not uniform, in size distribution. Both of these deviations tend to create smaller pores; hence, a larger ratio of d_g/d_p may apply and smaller particles may be strained.

Effect of Clinoptilolite Grain Size on Particle Release and Clogging. The fine clinoptilolite released more particles than the coarse clinoptilolite (Table 1). The abundance of particles on the grains appeared to be the same for the fine and coarse clinoptilolite. Assuming spherical grains and median grain diameters (d_{50}) estimated using the upper and lower sieve sizes (fine d_{50} = 0.58 mm; coarse d_{50} = 2.43 mm), the geometric specific surface area of the fine clinoptilolite

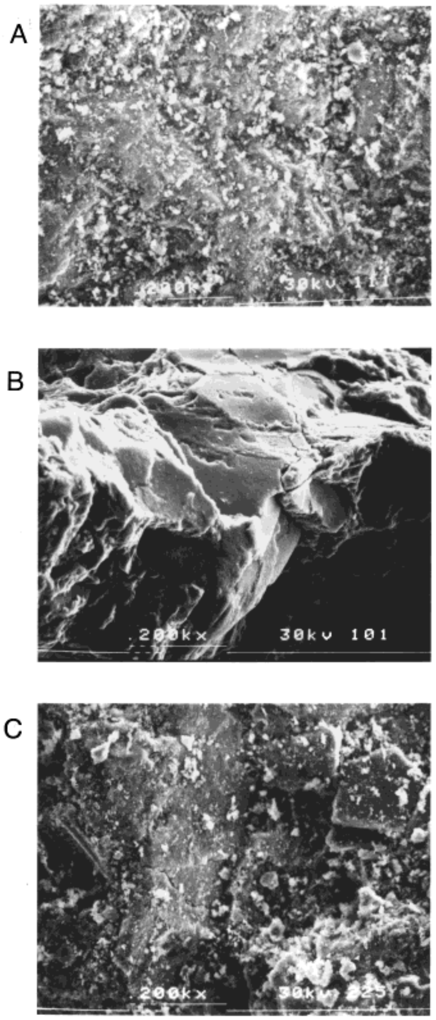

FIGURE 5. Scanning electron microscope images of grains from the (A) untreated, (B) rinsed, and (C) calcined coarse clinoptilolite. Rinsing removed the fine particles from the clinoptilolite surface; calcining did not. Grains were gold-coated; acceleration voltage was 30 kV. Magnification is 200 times. Scale (100 μm) shown by long bar below magnification (.200 kx).

(48 g cm^{-3}) is estimated to be 4 times greater than that of the coarse clinoptilolite (12 g cm^{-3}). In contrast, the fine clinoptilolite released only 1.1–1.7 times more particles than the coarse clinoptilolite. The amount of particle release is not simply related to the grain surface area, but we could not detect any other differences between the grains that might result in only slightly greater particle release for the fine clinoptilolite.

Clinoptilolite grain size indirectly affected the extent of clogging because the amount of particle release is related to the extent of clogging. For the untreated clinoptilolite experiments, greater particle release resulted in complete clogging in fewer pore volumes—4.3 for the fine clinoptilolite versus 5.4 for the coarse clinoptilolite (Table 1). For the rinsed clinoptilolite experiments, greater particle release resulted in a greater reduction in hydraulic conductivity ($K_{\min}/K_{\max}$)—0.42 for the fine clinoptilolite versus 0.60 for the coarse clinoptilolite.

Effect of Rinsing and Calcining on Particle Release. The two pretreatments, rinsing and calcining, reduced particle release and clogging. SEM examination indicates that rinsing removed a substantial fraction of the particles that were released from the untreated clinoptilolite (Figure 5). The reduction in the number of particles released reduced the extent of particle straining in the pores; hence, the hydraulic conductivity of the porous medium was better maintained. Similarly, other researchers (*18, 19, 30*) have shown that the extent of clogging depends on the clay- and silt-sized particles in the porous medium.

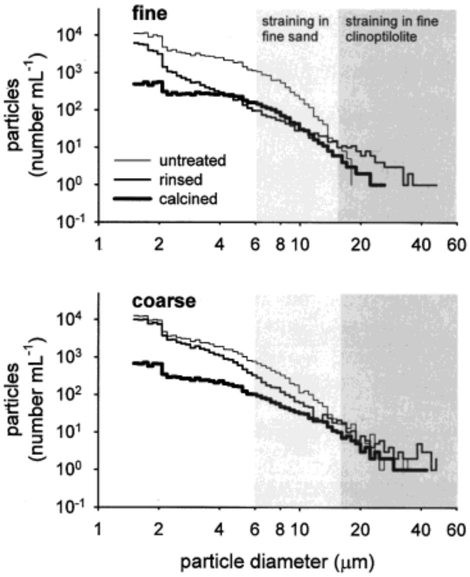

FIGURE 6. Particle size distributions for untreated, rinsed, and calcined clinoptilolite released from the clinoptilolite-only columns containing (top) fine and (bottom) coarse clinoptilolite. Measurements made at the maximum particle release concentration for each experiment. Particle size ranges capable of straining are shown for fine sand (light gray) and fine clinoptilolite (dark gray).

TABLE 2. Strontium Exchange by Untreated, Rinsed, and Calcined Versions of Fine and Coarse Calcium-saturated Clinoptilolite from a 1.0 × 10^{-3} M Strontium Chloride Solution at pH 5.6 ± 0.1

clinoptilolite grain size	clinoptilolite pretreatment	K_d, L kg^{-1} [a]
fine	untreated	7450 ± 540
	rinsed	4050 ± 220
	calcined	260 ± 20
coarse	untreated	3010 ± 310
	rinsed	1650 ± 130
	calcined	100 ± 12

[a] Distribution coefficients (K_d) presented as mean and standard deviation of 3 replicates.

Calcining improved hydraulic stability of the clinoptilolite not by reducing the abundance of fine particles but by increasing the strength of the clinoptilolite. SEM examination revealed roughly the same number of fine particles on the surfaces of the calcined and untreated clinoptilolite, but particle release was substantially reduced. The calcining temperature, 550 °C, is expected to drive off hydration water (*20*) but not to change the crystalline structure of clinoptilolite. Klieve and Semmens (*15*) showed that zeolites of greater strength (as measured by resistance to crushing) were less prone to attrition. Our crushing test showed that calcining elevated the clinoptilolite strength from the low end of the zeolite strength range to the high end. The relationship between crushing resistance and particle release is not clear, but it is reasonable to assume that the forces that oppose crushing are related to those that bind fine particles to grains. It is possible that driving off hydration water allows particles to bind together more strongly by shortening their separation distance.

Effect of Grain Size and Pretreatment on Strontium Sorption. The fine clinoptilolite adsorbed strontium about 2.5 times as strongly as the coarse clinoptilolite, a difference that may be related to the difference in surface area. The geometric surface area of the fine clinoptilolite is about 4 times greater than that of the coarse clinoptilolite, fairly close to the difference in K_d. Alternatively, K_d may be related to the volume and, hence, the mass of the particles, because a large fraction of the cation binding sites are located in internal pores; however, the kinetics of cations reaching these internal pores is quite slow relative to the initial exchange at surface sites (*36*, *37*).

Rinsing the clinoptilolite grains resulted in a decrease in K_d by a factor of 1.8 for both the fine and coarse clinoptilolites. We know that rinsing removed most of the fine particles from the surfaces of the clinoptilolite grains (Figure 5); therefore, the overall surface area of the clinoptilolite must have decreased. The surface area of these fine particles is not accounted for in the geometric estimates of surface area used to compare the effect of grain size on particle release and K_d. Calcining reduced the strength of strontium sorption by a factor of about 30 relative to the untreated clinoptilolites. We hypothesize that access to internal porosity and adsorption sites was restricted by calcining.

Implications for Use in Permeable Reactive Barriers. This research identified three important factors for judging the suitability of clinoptilolite as a permeable reactive barrier material: groundwater composition and ionic strength, porous medium grain size, and type of clinoptilolite pretreatment. Low ionic strength, sodium-dominated groundwater may cause particle release, which may result in clogging. Because the ionic strength and calcium content of the West Valley is relatively high, particle release from the clinoptilolite in the pilot barrier is not expected. Groundwater ionic strength must be considered in transferring this technology to other sites.

If particles are released from a clinoptilolite barrier, the grain size of the porous medium down-gradient of the barrier will determine the extent of clogging. Assuming that straining is the main mechanism of particle release, we expect that clogging may occur in a fine- to medium-grained sand but not in a coarse sand. Poorly sorted sands may be more susceptible to clogging than well-sorted sands. Also, it should be noted that sands coated by positively charged minerals (e.g., ferric oxyhydroxides) will promote deposition of negatively charged clinoptilolite particles, which may accelerate clogging or allow clogging to occur in a sand that does not strain particles.

The calcining pretreatment virtually eliminated particle release and clogging but at the expense of strontium adsorption. Rinsing reduced particle release and clogging by about a factor of 1.2−2.0 at the expense of a factor of 2.5 reduction in strontium adsorption. Another consideration in assessing the suitability of a pretreatment is the cost of performing these pretreatments on a large scale. One of the main reasons that clinoptilolite was chosen as a candidate material was high “cation exchange capacity per dollar” (*12*). Pretreated clinoptilolites would have to be reexamined from this perspective after the cost of large-scale pretreatments was added.

Finally, the combination of particle release and strong adsorption of strontium suggests that colloid-facilitated transport of strontium may be a problem down-gradient of the barrier (*38*). The potential of two zeolite minerals in the volcanic tuff at Yucca Mountain, clinoptilolite and heulandite, for facilitating the transport of radionuclides have been investigated (*39*, *40*). Assuming a steady-state particle concentration of 1 mg L^{-1}, a conservative K_d of 1000 L kg^{-1} (*1*, *13*), and groundwater contaminated with 1 nM ^{90}Sr, particles saturated with ^{90}Sr will carry 10^{-12} M ^{90}Sr. This concentration of ^{90}Sr would emit 12 000 pCi L^{-1} of beta radiation, in excess of the U.S. Environmental Protection Agency’s maximum contaminant level for beta-emitting radionuclides (8 pCi L^{-1}). Assuming that the kinetics of strontium desorption from the clinoptilolite are sufficiently slow to allow colloid-facilitated transport to occur and that the clinoptilolite particles are mobile in the porous medium, colloid-facilitated transport of strontium could be a problem in clinoptilolite barrier installations.

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