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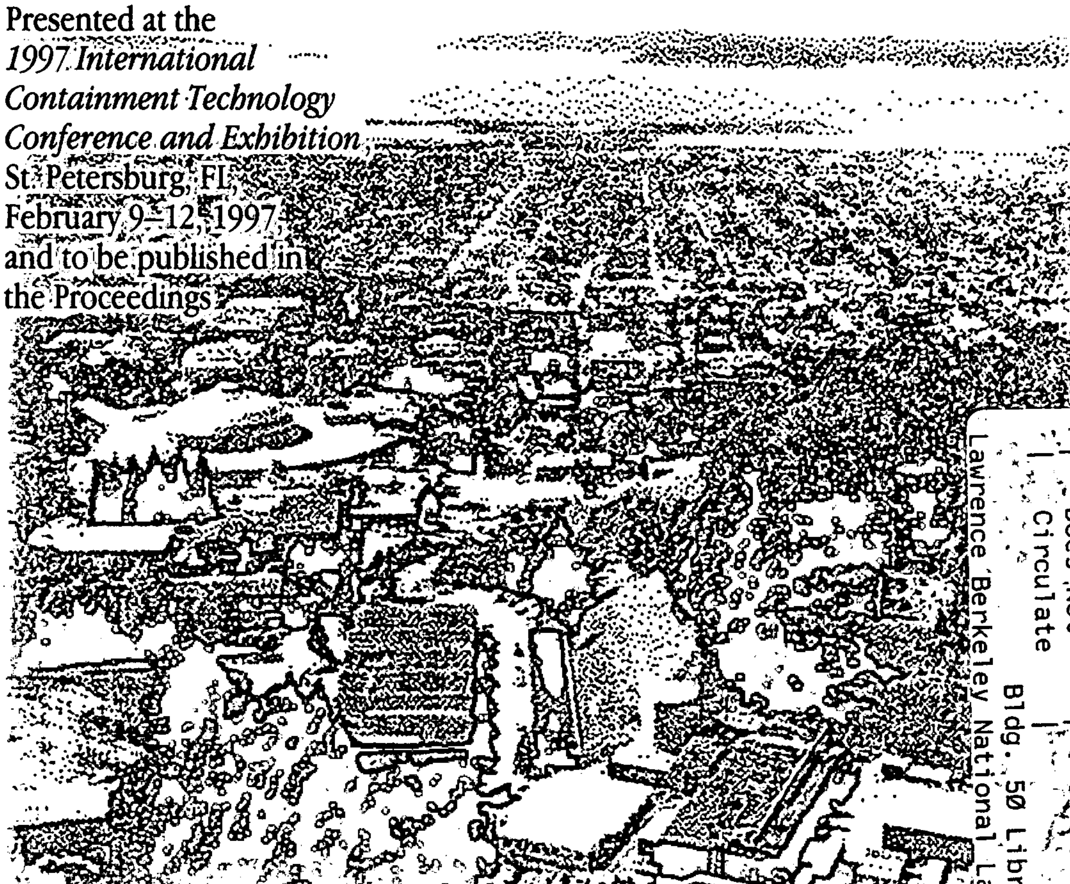

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EFFECT OF DILUTION AND CONTAMINANTS ON STRENGTH AND HYDRAULIC CONDUCTIVITY OF SAND GROUTED WITH COLLOIDAL SILICA GEL

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INTRODUCTION

Colloidal silica (CS) is a low-viscosity liquid that can be made to gel by addition of brine. This property allows it to be injected into, or mixed with, soil, so that after gelling the colloidal silica blocks the pore space in the soil and forms a barrier to the flow of contaminated groundwater or non-aqueous liquids (NAPLs). Gelled-in-place CS was first studied for the petroleum industry (Jurinak *et al.*, 1991, Seright 1993) and later for protecting groundwater quality (Yonekura et al 1992, 1993, Noll *et al.* 1992, Persoff *et al.* 1994, 1995, Moridis *et al.* 1996). Noll (1992) investigated the use of colloidal silica diluted so that its solids content was reduced from 30% (a typical nominal value for material as delivered) to values as low as 5%. The more dilute colloids could still be made to gel, although more slowly, and the resulting gel was weaker. Because the proposed application of colloidal silica grout involves emplacing it in the subsurface by permeation, jet grouting, or soil mixing where its role as a barrier will be to resist flow of contaminants, the effects of these contaminants on the properties of the grouted soil is also of interest.

This work comprised four tasks. In Task 1, samples of grouted sand were prepared with a range of CS dilutions, for measurement of hydraulic conductivity and unconfined compressive strength. In Task 2, these properties were measured on samples of grouted sand that incorporated 5% volumetric saturation of NAPLs. In Task 3, samples, prepared without any contaminants, were immersed in contaminant liquids and tested after 30 and 90 days.

Task 4 was added because NAPL contamination in the samples of Tasks 2 and 3 impelled modifications in the test methods, and comparison of the results of Task 2 and Task 1 suggested that these modifications had introduced errors. In Task 4, samples were tested both ways, to confirm that in Tasks 2 and 3 strength was underestimated and hydraulic conductivity was overestimated. Despite the existence of these known systematic errors, the inclusion of control samples in Tasks 2 and 3 permits conclusions to be drawn from these data.

MATERIALS

The CS used in this work was DuPont Ludox SM, with 29.5 weight percent silica. The sand was Monterey #0-30 sand, a silica sand. Brines were made from distilled water and reagent NaCl, and distilled water was used for dilutions. pH adjustment was done by titration with concentrated HCl.

METHODS

SAMPLE PREPARATION

Task 1: Uncontaminated Monterey sand grouted with five dilutions of CS.

First the grout was prepared and then it was combined with the soil or sand in a mold to make each sample. The CS was first diluted from its as-received silica concentration of 34.3 wt % with distilled water. For grouts that required pH adjustment, the pH was then lowered to approximately 8.5 by titration with concentrated HCl. Next, NaCl brine was added with constant swirling of the diluted colloid, and final pH adjustment was done. pH adjustment was done in two stages to minimize the time needed to adjust pH after the brine was added.

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Three hundred forty g of sand and 78 mL of grout were mixed to fill each 5 cm diameter x 10 cm long (2 x 4 inch) cylinder mold. The bottoms of the plastic cylinder molds were removed and replaced by caulked-on lids; this allowed the samples to be slid out of the molds for testing. To prepare each sample, the liquid grout was poured into the mold, and the pre-weighed sand was poured slowly into the liquid while gently shaking the mold to settle the sand. These amounts of sand and grout were found to fill the mold without excess liquid or solid.

Table 1. Formulae for Task 1 samples.

Volume liquid per sample (mL)	Volume colloid per sample (mL)	Silica concentration after dilution (wt %)	Added brine [NaCl], Molarity	volume brine per sample (mL)	final [NaCl], Molarity	volume distilled H_2O	mass sand per sample (g)	final pH
78	13	4.9	1.8	13	0.3	52	340	6.95
78	20	7.4 [a]	1.2	13	0.2	46	340	6.95
78	26	9.8	1.2	13	0.2	39	340	6.48
78	52	19.7[a]	0.6	26	0.2	0	340	10 (not adjusted)
78	71.5	27.0	1.2	6.5	0.1	0	340	10 (not adjusted)

[a] these formulae also tested in Tasks 2 and 3

Task 2: Monterey sand, contaminated with NAPL, grouted with two dilutions of CS.

The data of Wilkins *et al.* (1995) suggest that in the NAPL-contaminated unsaturated zone of sandy soil, volumetric saturation of water and NAPL are typically 10 and 5 % of pore space. Samples for Task 2 were made as for Task 1a, except that the sand was prepared by addition of first water, and then NAPL, to produce these saturations in the sand before the sand was poured into the mold. The dilution water in the grout was reduced to compensate for the water pre-added to the sand. Three NAPLs were used in this task: C_2Cl_4 (perchloroethylene, PCE), CCl_4, $C_6H_5NH_2$ (aniline). Samples were also prepared with 10% water saturation and no NAPL.

Task 3: Uncontaminated Monterey sand grouted with two dilutions of CS, immersed in contaminants.

Samples were prepared as for Task 1, and then immersed in one of nine liquids: the three NAPLs, water saturated with each of the three NAPLs, water saturated with an equimolar mixture of the three NAPLs, HCl diluted to pH 3, and distilled water.

TEST METHODS

Measurement Of Unconfined Compressive Strength.

Unconfined compressive strength was measured by ASTM C-39-86, using a loading rate of 50 lb./min. In this test, flat and parallel sample ends are assured by capping the ends. For Task 1, the samples were capped with Cylcap sulfur mortar, according to ASTM C-617-72 To avoid exposing personnel to NAPLs during the capping and testing, different capping and testing procedures were used for Tasks 2 and 3. Hydrostone, a gypsum plaster, was used so that the capping could be done in a hood without exposing the samples to heat. Also, after capping, the samples were enclosed in zip-lock plastic bags and tested in the bags.

Measurement Of Hydraulic Conductivity.

Hydraulic conductivity was measured in a flexible wall permeameter by ASTM D-5084, at a net confining pressure of 207 kPa (30 psi). As with the strength measurements, safety considerations required that the technique be modified for Tasks 2 and 3. Sample preparation was designed to prevent the latex membrane from contacting the solvent and also to prevent any evaporation of the solvent. To prevent the latex membrane from contacting the solvent, 0.076-

mm (0.003-inch-) -thick Teflon sheet, (Boart Longyear, Salt Lake City) was wrapped around the sample with a 1-cm overlap, held in place with vacuum grease. More details are presented elsewhere (Persoff *et al.* 1997).

RESULTS

Task 1: Effect of dilution on strength and hydraulic conductivity

The compressive strength and hydraulic conductivity of Monterey sand grouted with CS are shown in **Figure 1.** The sample consolidates to some degree during measurement of hydraulic conductivity under 207 kPa (30 psi) confining pressure. The volume of each sample was measured both before and after measurement of hydraulic conductivity, and finally the total solids were determined by drying the sample. From these data the dry density and (assuming a density value for the solids) the porosity before and after consolidation was calculated. These porosity data are shown in **Figure 2.** The difference between the initial and final porosity is a measure of sample consolidation that occurs when the sample is subjected to the confining pressure; the confining pressure caused minimal consolidation of the grouted Monterey sand.

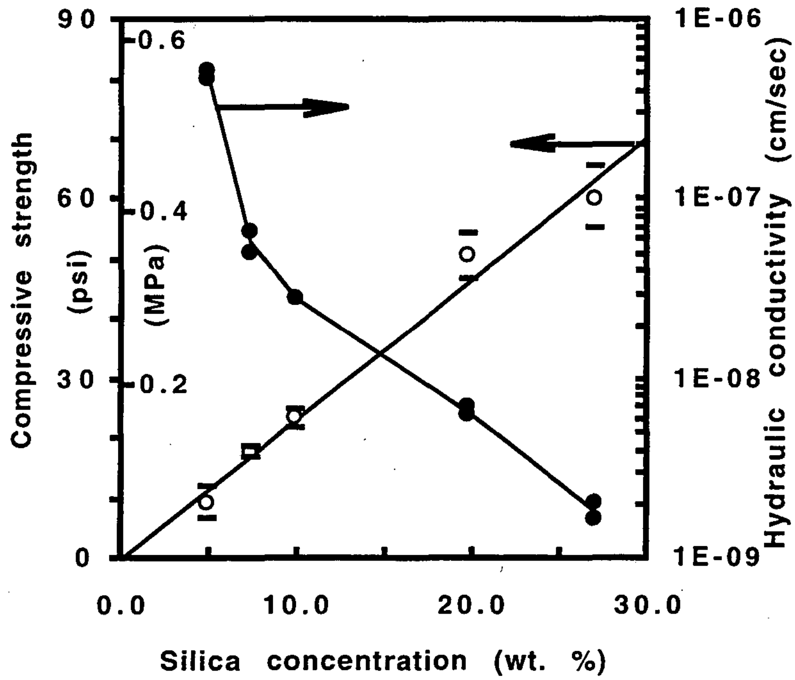

Figure 1. Compressive strength and hydraulic conductivity of samples of Monterey sand grouted with various dilutions of Ludox SM. Error bars are standard deviation of five strength measurements; duplicate hydraulic conductivity measurements are shown. The strength line is a least-squares fit, not forced through the origin.

Tasks 2 and 3: Effect of inclusion of NAPLs on strength and hydraulic conductivity.

The results of measurements on Task 2 and 3 samples are shown in **Tables 2 and 3.**

DISCUSSION

Strength of Task 1 samples.

Figure 1 shows that as the silica content of the grout is reduced by dilution, the strength of the sample decreases and the hydraulic conductivity increases. The Monterey sand itself is not cohesive, and any strength results from the cementing effect of the grout, which increases linearly with the amount of silica in the grout. This suggests that the colloidal particles bond not only to each other but also to the silica surface of the sand.

The strength of sand grouted with colloidal silica and sodium silicate-glyoxal grouts was investigated by Yonekura and Miwa (1993). They found that the strength of sand grouted with colloidal silica was independent of gel time in the range 10 sec - 1 hr, and continued to increase during 1000 days of aging. Tested at 100 days, the strength was 95 psi, and the ultimate strength was more than twice that.

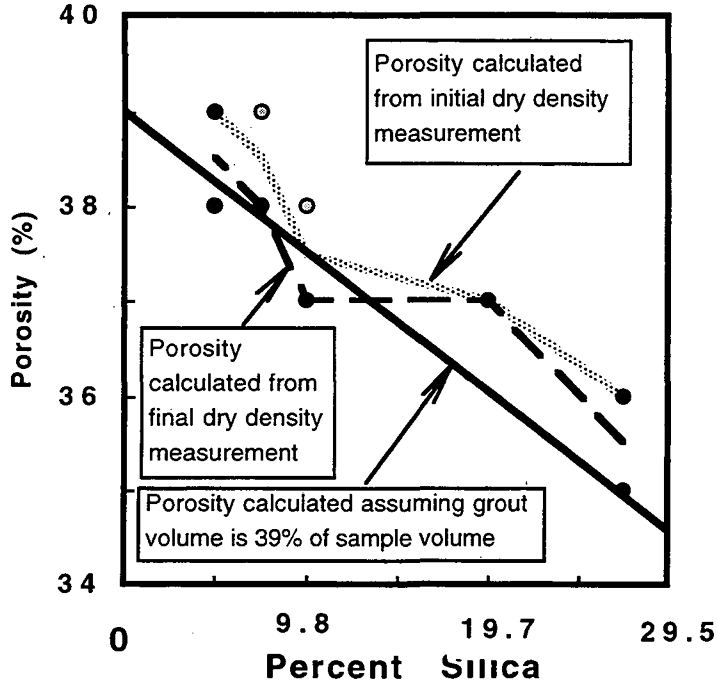

Figure 2. Porosity of Task 1 samples before and after consolidation.

Table 2. Compressive strength and hydraulic conductivity of samples of Monterey sand, contaminated with NAPLs and grouted with two dilutions of CS (Task 2).

percent silica	contaminant	unconfined compressive strength (kPa) mean of 5	std. dev	hydraulic conductivity (cm/sec) mean of 2	difference between 2
7.4	PCE	149.9	8.14	3.30E-07	1.00E-07
	CCl_4	129.3	8.96	7.95E-07	3.10E-07
	aniline	129.7	10.7	2.80E-07	1.00E-07
	water	128.0	9.03	2.55E-07	1.50E-07
19.7	PCE	273.9	15.4	1.35E-08	1.00E-09
	CCl_4	296.3	21.7	4.25E-08	2.10E-08
	aniline	236.3	20.8	1.33E-08	7.50E-09
	water	295.9	36.0	2.40E-08	1.60E-08

Table 3. Unconfined compressive strength (kPa) of samples immersed in various liquids (Task 3).

percent silica	not immersed (Task 1) mean of 5	std dev	immersion liquid	30 day immersion mean of 3	std dev	90 day immersion mean of 3	std dev
7.4	123.9	7.24	water	181.8	17.6	165.7	4.76
			PCE	186.9	13.9	168.2	8.55
			CCl_4	170.9	17.5	169.0	2.90
			aniline	93.29	18.1	116.3	3.10
			water s/w PCE[b]	189.3	10.5	179.6	12.6
			water s/w CCl_4	201.5	12.1	169.8	10.8
			water s/w aniline	161.3	1.10	138.2	23.2
			water s/w mix of 3	174.5	14.0	170.1	6.55
			pH 3	166.1	11.1	155.8	6.55
19.7	349.6	26.4	water	492.0	21.2	533.8	55.4
			PCE	493.8	6.55	567.3	6.14
			CCl_4	499.7	3.38	535.1	11.1
			aniline	327.0	41.7	355.6	27.5
			water s/w PCE	456.9	38.2	533.3	29.6
			water s/w CCl_4	463.8	19.8	553.0	33.8
			water s/w aniline	497.5	41.5	413.4	22.1
			water s/w mix of 3	501.5	38.1	587.1[a]	0.00[a]
			pH 3	531.9	24.3	601.4[a]	2.21[a]

[a] mean and difference between two samples.
[b] s/w = saturated with

Hydraulic conductivity of Task 1 samples.
The hydraulic conductivity of the grouted sand or soil is the principal property of interest. Generally a value of 10^{-7} cm/sec is taken as the criterion for acceptable barrier material. The data in Figures 3 and 4 show that this criterion is met by all the samples made with at least 7.4 % silica. This number represents therefore the lower limit for dilution of Ludox SM for use as a barrier material.

Figure 1 shows a strong effect of the silica concentration on the permeability of the Monterey sand samples. The hydraulic conductivity of the grout in these samples was unaffected by consolidation. The sand grains themselves are effectively impermeable and the measured hydraulic conductivity can be understood to represent the hydraulic conductivity of the gelled Ludox SM itself, multiplied by a factor of approximately 0.38, representing the volume fraction of the sample occupied by gelled grout.

The relationship between silica concentration and hydraulic conductivity can be explained, at least qualitatively, by considering the structure of the gelled grout. By volume, the gelled grout is mostly water, and the space between the gelled chains of silica particles constitutes a network of pores through which water can flow. The low value of hydraulic conductivity results from a highly divided flow path with many small pores. At 27.0 % silica the grout is (100 - 27.0/2.65) = 90% water by volume, and at 7.4 % it is about 97% water. This 8 % increase in flow area is too small to account for a 30-fold increase in the hydraulic conductivity. Flow resistance results from viscous drag on water as it flows through a tangle of chains of gelled particles. Increasing the silica concentration by a factor of (27.0/7.4)=3.67 reduces the space between chains. This space between chains may be considered as a measure of the effective radius of pores. Approximate the pores as Hagen-Poiseuille flow in parallel tubes: for fixed pressure gradient and viscosity, the flow through each tube is proportional to the fourth power of the radius. Flux, or Darcy velocity, is thus proportional to the square of the radius. While the geometry of the system is not defined well enough to permit actual calculation of the change in permeability, the effect of reduced friction drag by chains of silica particles can account for the increase in permeability.

Effects of test-method modifications for Tasks 2 and 3
Three groups of samples included in Tasks 1, 2, and 4 were tested by both unmodified and modified methods. Matched data in Table 4 show that the samples of Tasks 1 and 2, identical in composition but differing in the test method, gave different results. This suggested that the modifications (introduced because of NAPL contamination) had the effect of increasing the measured hydraulic conductivity and decreasing the measured strength. To confirm this, in Task 4, additional samples were prepared without contamination, and tested by both methods. The results of these tests are summarized in Table 4.

Table 4 shows that the modified test method for strength causes an underestimate of strength, but only for the stronger samples (19.7 and 27.0 % silica). This is reasonable because the requirement for the capping compound is that it not fail before the sample. Similarly, (although here the small number of samples makes the conclusion less certain) the use of Teflon sheet caused an overestimate of the hydraulic conductivity. In the light of these results, we caution that the results of Tasks 2 and 3 can be interpreted only to determine the effects of inclusion or immersion in contaminants *relative to the water control.*

The use of Teflon sheet or tape (with some variation as to materials and method of wrapping) to protect latex membranes appears to be the standard method for preparing contaminated samples for ASTM D-5084 (Daniel and Trautwein, 1994). In Task 4 (Table 4), we treat the overestimate in hydraulic conductivity as an additive factor representing a parallel flow path. For a 5-cm diameter sample (and at 200 kPa confining pressure) this wall effect is equivalent to 3.4E-8 or 3.0E-8 cm/sec hydraulic conductivity. This degree of error becomes significant when measuring samples as tight as these.

Table 3 shows that samples gained strength during the immersion, except for samples immersed in aniline and water saturated with aniline. In that sense, immersion in aniline weakened the samples.

Table 4. Comparison of values measured with modified and unmodified test methods.

Property	% silica	ASTM	Task 1 not modified			Task 2 modified			Task 4 not modified			Task 4 modified		
			n	mean	s**	n	mean	s**	n	mean	s**	n	mean	s**
Unconfined compressive strength(kPa)	7.4	C-39	5	123.9	7.24	5	128.0	9.0						
	19.7	C-39	5	349.6	26.4	5	295.9	36.0						
	27.0	C-39	5	416.4	35.1				4	416.1	18.6	3	367.5	9.7
Hydraulic conductivity (cm/sec)	7.4	D-5084	2	4.95E-08	1.7E-08	2	2.55E-07	1.5E-07						
	19.7	D-5084	2	6.65E-09	7.0E-10	2	2.40E-08	1.6E-08						
	27.0	D-5084	2	1.9E-9	0.4E-9				1	5.0E-9		1	3.9E-8[a]	

[a] after this measurement, the sample was remeasured using the unmodified method, and hydraulic conductivity was 9E-9 cm/sec.
**standard deviation, or difference between 2 measurements

CONCLUSIONS

1. The unconfined compressive strength of sand grouted with Ludox SM is proportional to the concentration of colloidal silica particles, up to a maximum of approximately 400 kPa (60 psi).

2. The hydraulic conductivity of sand grouted with Ludox SM decreases with increasing concentration of colloidal silica particles. For silica particle concentration greater than 7.4 % by weight , the hydraulic conductivity is less than 1.0 x 10^{-7} cm/sec; that is, it meets the generally accepted criterion for a barrier material. In this range, the log of hydraulic conductivity decreases approximately linearly with increasing concentration of colloidal silica particles.

3. Monterey sand provided a skeleton to prevent consolidation of grout under confining pressure. Under these conditions measured k is therefore a function of the grout, and variation of k with Si content can be explained on the basis of flow through a network of Si chains.

4. Samples immersed in water continued to gain strength for 95 days. Immersion of samples for up to 95 days in aniline, or in water saturated with aniline, weakened the samples. Immersion for up to 95 days in the other test liquids (PCE, CCl_4, water saturated with these NAPLs, water saturated with the mixture of 3 NAPLs, and HCl diluted to pH 3) had no statistically significant effect, i.e., they gained strength during 95 days of immersion the same as those immersed in water.

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