GEOTECHNICAL METHODS FOR COLLOIDAL CONTAMINANT TRANSPORT

METHODES GEOTECHNIQUES POUR LE TRANSPORT DE CONTAMINANT COLLOIDAL

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SYNOPSIS: Methods to transport colloidal contaminants in the subsurface are of paramount importance in remediation. In an experimental study, the feasibility of mobilizing colloids using sub- and ultra-sonic vibrations was investigated. ~~Theoretical~~ considerations involved in the mobilization of colloidal particles were examined. Subsonic vibrations in soils contaminated with Non-Aqueous Phase Liquids (NAPLs) resulted in upto 85% recovery of residual NAPL colloids. Ultrasonic vibrations in clayey soils mobilized particles smaller than 0.04 mm and fractured particles in the size range of 0.04 mm to 1 mm. The results from the laboratory tests, in general, indicate the potential use of vibratory methods in in-situ remediation.

INTRODUCTION

In-situ treatment of contaminated sites has been a subject of active research in recent years. An important problem concerning in-situ remediation is the transport of colloidal contaminants in the subsurface. Transport of contaminants in ground water is generally treated by partitioning the contaminant between the mobile aqueous phase and the immobile solid phase. However, it is also recognized in several studies (McDowell-Boyer, et al. 1986; Huling, 1989; for example) that small solid particles and macromolecules which exist in some subsurface environments, act as mobile sorbents as they travel in the aqueous phase. The small solid particles which are in the clay and colloidal size ranges, have high surface areas per unit mass, thus posing a significant sorption potential. The migration of contaminants in association with the colloids is not well understood at the present time and is generally neglected in modeling contaminant migration.

Colloids are of two groups: organic and inorganic. Organic colloids include bacteria, humic substances, and non-aqueous phase liquids (NAPLs). Clay particles and metal oxides form the inorganic colloids. The present study focuses on vibratory transport of NAPL and clay colloids. Subsonic and ultrasonic vibrations were used to investigate the mobility of NAPLs and clay particles respectively. Theoretical considerations pertaining to the mobility of the two types of colloids are first examined prior to the discussion of experimental investigations.

THEORETICAL CONSIDERATIONS

Figure 1 illustrates the principal forces involved in the kinetics of colloidal particle deposition in porous media. Other physical processes such as Brownian diffusion may also be included in colloidal transport analyses. The chemical interactive forces (F_{chem} in Fig. 1) include all processes arising from the electrostatic, hydrophobic and other specific chemical characteristics of the colloidal particle and the porous media surfaces. One of the earlier quantitative expressions for the kinetics of colloidal particles was given by Yao et al. (1971):

$$\frac{dC}{dL} = \frac{-3}{4} \alpha_{p.c} \eta_{p.c} \frac{(1-\varepsilon)}{a_c} C \qquad (1)$$

where C is the concentration of particles of a specific colloidal size, L is the length of travel of the particles in the porous media, a_c is the radius of the grains (assumed to be spherical) comprising the media, and ε is the porosity of the media. $\alpha_{p.c}$ and $\eta_{p.c}$ are the kinetic coefficients describing chemical and physical processes involved (Fig. 1) in the deposition process of colloidal particles (c) on stationary porous media particles (p). Empirical expressions for the determination of these coefficients have been given by Rajagopalan and Tien (1976).

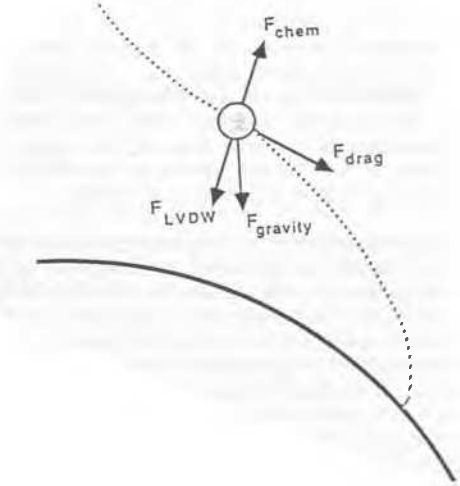

Fig. 1. Forces acting on a colloidal particle in the porous media $F_{gravity}$ = gravity force F_{drag} = drag force; F_{LVDW} = London-van der Waals force, F_{chem} = chemical forces. [Adapted from Tobiason and O'Melia (1988)]

Transport of solitary NAP colloids, on the other hand, depends on a host of parameters related to soil-NAPL characteristics and the geometry of the throats in which they reside. Once an oil ganglion is set in motion, its subsequent fate depends largely on the geometry of the track downstream in the direction of the macroscopic pressure gradient. In one of the most comprehensive studies, Payatakes et al. (1980, 1982) developed mobilization criteria for NAPL ganglia using a constricted-tube network model to simulate the porous media (Fig. 2).

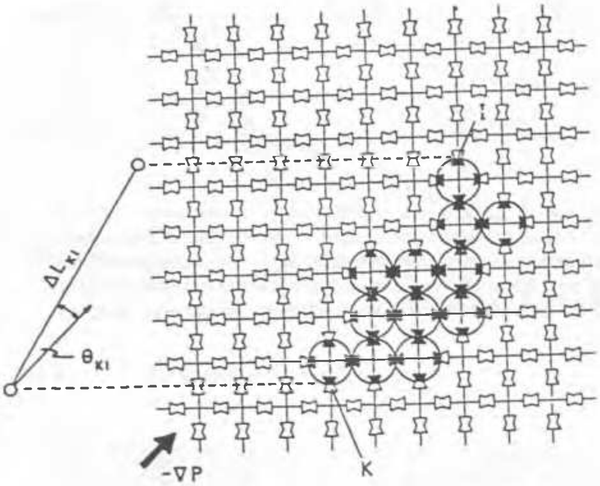

Fig. 2. Payatakes' Constricted-tube Network Model. Circles represent conceptual elemental void spaces. An idealized oil ganglion occupying twelve adjoining pores is also shown. From Payatakes et al. (1982).

From equilibrium conditions at the interfaces, the mobilization criterion for the ganglion between any two throats 'K' and 'I' (Fig. 2) was obtained as

$$\left(\frac{\mu_w V_f}{\gamma_{ow}}\right) > \left(\frac{k_{rw} k}{\beta_{kI}}\right) \qquad (2)$$

where μ_w = dynamic viscosity of the aqueous phase, V_f = superficial velocity of the aqueous phase, γ_{ow} = interfacial tension, k_{rw} = relative permeability to water, and k = absolute permeability. β_{KI}, known as the appendix mobility factor, is a function of geometrical parameters of the void space in the porous media. Simplified versions of Eq. 2 have been used in various studies including Wilson et al. (1984) and Hunt et al. (1988).

The effect of subsonic vibrations in a saturated porous medium is to momentarily increase the pore pressures resulting in an increase in V_f (Eq. 2), thereby enhancing the mobility of solitary NAPL ganglia. The effect of ultrasonic vibrations in the porous media, on the other hand, is to cause cavitation which is a rapid and repeated formation, and resulting implosion of microbubbles in the aqueous phase. The number of cavitation bubbles collapsing per second may be in the millions; hence their cumulative effect can be sufficient. In a porous media, these cavitation bubbles generate high differential fluid-particle velocities and are capable of dislodging micron-size colloidal particles in the system by overcoming the physical forces (Fig. 1). Results from experimental investigations described in the following section, reflect these implications.

EXPERIMENTAL PROGRAM AND RESULTS

The purpose of the experimental program was to create sub- and ultra-sonic vibrations in laboratory soil columns and evaluate the feasibility of mobilizing colloids. The laboratory tests were intended to be exploratory in nature; consequently, not all in-situ conditions could be simulated. Costs of remediation of contaminated soils limited the sizes of soil columns used in the experiments. The following sections include description of the experimental program and discussion of results.

Effect of subsonic vibrations on NAPL colloidal transport

Experiments were conducted using a commercially available concrete vibrator (10,000 to 12,000 revolutions per minute) in PVC columns (6 ft. high and 1 ft. diameter) filled with sandy soils. The vibrator was housed in a well chamber and an injection chamber as shown in Fig. 3. Water was pumped into the injection chamber through a metal tube. Several jet ports were located to facilitate waterflooding. The dislodged ganglia were pumped out from the well chamber with the help of a withdrawal pump. Two types of soils were chosen; #0 soil with 90% retention between US sieves 20 and 60 and #1 soil with 90% retention between US sieves 10 and 40. NAPLs chosen for the tests were No. 2 heating oil and jet fuel. The soil in the column was saturated with water before introducing a known volume of NAPL at the top. The NAPL was allowed to drain from the bottom and the soil column was backflooded with water to extract dislodged NAPL. The soil column was then vibrated, simultaneously maintaining a flow of water. The percentage of NAPL quantity mobilized due to vibrations was obtained using mass balance.

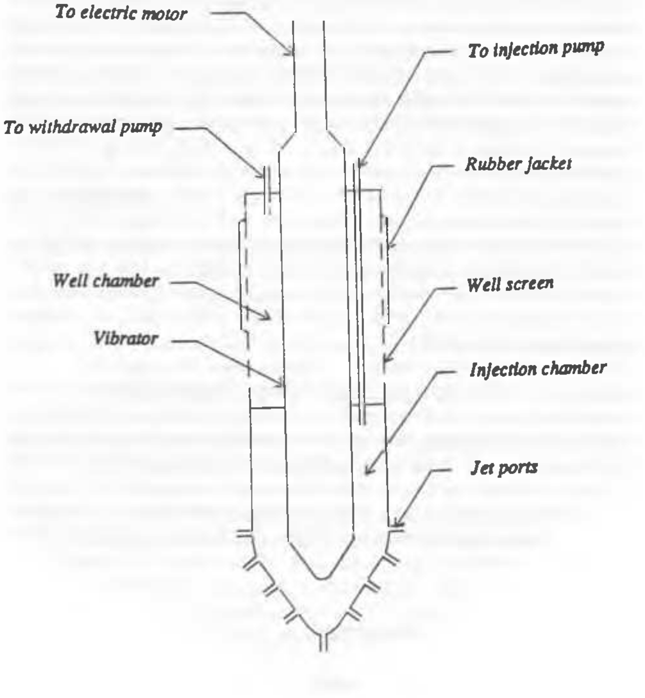

Fig. 3. Design of vibroflot used in the experiments.

In test series 1 to 4 (Table 1), experiments were first conducted without vibrations and with only waterflooding provided by flow rates of 2.0 gpm (1.5 x 10^{-4} m^3/sec). Insignificant NAPL quantities were recovered in these experiments. When the same experiments were conducted with waterflooding augmented by vibrations, upto 52% of NAPL was mobilized and recovered. Test series 5 and 6 which were carried out with vibrations and flow rates of 4.0 gpm (3.0 x 10^{-4} m^3/sec), showed improved separation and mobilization of NAPL ganglia. Several parameters were identified to be controlling the process: i) type of soil and NAPL, ii) flow rates in the column, and iii) time of treatment. In summary, the results show that the process is more efficient in coarse soils than in fine soils. The flow rates of water in the column seem to directly influence the break-up of ganglia. The results also show that longer treatment time yields no significant improvement in recovery. Multiple runs resulted in further recovery of NAPL; however, the improvement in recovery was reduced with each repetition of the process as a result of soil compaction.

Table 1. NAPL Colloidal transport due to vibrations

Test #	Soil Type	NAPL Type	% of NAPL mobilized
1	#0	#2 heating oil	28.5%
2	#0	jet fuel	42.4%
3	#1	#2 heating oil	52.0%
4	#0	#2 heating oil	28.2%
5	#1	#2 heating oil	73.9%
6	#0	jet fuel	85.0%

Due to limitations of the experimental set-up, a fundamental understanding of the effect of important parameters such as vibration frequency could not be obtained. The experimental set-up also prevented a study of attenuation of vibrations in the soil-NAPL-water medium and an assessment of radius of influence of the vibratory phase separation. The nature of vibrations generated in the column could not be characterized due to reflection and refraction effects from the walls of the column. The results shown in Table 1 should therefore be viewed as preliminary and should not be used to extrapolate to a semi-infinite medium.

Effect of ultrasonic vibrations on clay colloidal transport

Experiments were conducted using a probe-type Ultrasonic Liquid Processor in plexiglass columns (1.5 ft. high and 1 ft. diameter) filled with mixtures of commercial sand and natural New Brunswick clay. A power supply converts conventional 50 or 60 Hz alternating current to 20 KHz electric energy which is then fed to a convertor in the horn, where it is transformed to mechanical excitation. A maximum tip amplitude of about 60 microns was achieved with this horn with a corresponding power requirement of 500 watts.

Mixtures of commercial sand and natural New Brunswick clay were compacted in the plexiglass columns at desired densities using standard proctor hammer. Grain-size distributions of representative samples were obtained by conducting sieve and hydrometer analyses in accordance with ASTM D422. The soil column was saturated by pumping water from bottom of the sample. The ultrasonic probe was then inserted with minimal excitation (to facilitate the insertion). Saturated hydraulic conductivities of the soils were measured before and after ultrasonic treatment using the falling-head method. Under the heads used in the process of permeation, no clay particles were observed in the effluent water. The treatment was then conducted incrementally with hydraulic conductivity measured at the end of every time increment. At the end of each test, representative soils samples at three depths in the column away from the probe were tested for grain size distribution and hydrometer analyses. Hydrometer analyses were also conducted on the effluent slurries obtained during treatment.

The change in grain size distribution curves of the soil as a result of ultrasonic excitation is presented in Fig. 4 for a representative test. Figure 4 shows that ultrasonic excitation reduced the percentage of fine particles thereby causing an increase in hydraulic conductivities. The grain-size distribution curves also show that, after the ultrasonic treatment, a decrease in fine particle percentage did not result in a corresponding increase in coarser fractions. This finding is attributed to the abrasion and/or fracturing of sand-sized grains resulting from ultrasonics. Results from hydrometer analyses of the effluent slurry are also shown in Fig. 4. These indicate that the maximum size of the particles mobilized by ultrasonics was about 0.04 mm and that particles in the size range of 0.04 mm to 1 mm were subjected to fracturing. An increase in hydraulic conductivity was observed in all the tests with a maximum being reached after about 8 to 10 minutes of excitation.

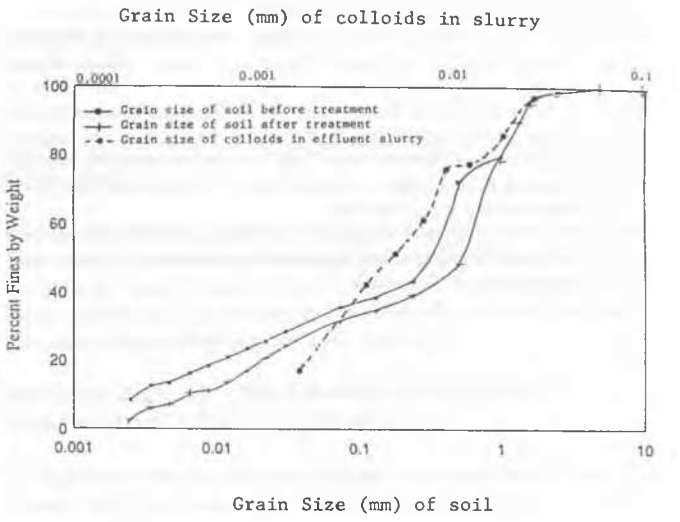

Fig. 4. Grain Size Distribution of Soils and Effluent Slurry in an Ultrasonic Test.

CONCLUSIONS

Laboratory tests were conducted to evaluate the feasibility of mobilizing organic and inorganic colloids using sub- and ultra-sonic vibrations. Results show that subsonic vibrations mobilized upto 85% of NAPL colloids in laboratory columns. Important parameters governing the mobilization are: i) type of soil and NAPL, ii) flow rates in the soil column, and iii) time of treatment. Ultrasonic vibrations mobilized clay colloids smaller than 0.04 mm and subjected particles in the size range of 0.04 mm to 1 mm to fracturing. Due to limitations of the experimental set-up, the zones of influence of vibratory mobilization could not be obtained. Boundary effects due to reflection and refraction of waves from the walls of the laboratory columns need to be addressed before designing vibratory methods for in-situ implementation. Economic feasibility of these methods depend on the attenuation characteristics of vibrations in contaminated soils. Future studies may focus on these aspects.

ACKNOWLEDGMENTS

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REFERENCES

Huling, S.G., (1989). *Facilitated Transport*. Superfund Ground Water Issue, EPA/540/4-89/003, Pp. 1-5.

Hunt, J.R., Sitar, N. and Udell, K.S., 1988. Non-Aqueous Phase Liquid Transport and Cleanup, I. Analysis of Mechanisms, Water Resources Research, Vol. 24, No. 8, Pp. 1247-1258.

McDowell-Boyer, L. M., Hunt, J.R., and Sitar, N., (1986). Particle Transport Through Porous Media. Water Resources Research, Vol. 22, No. 13, Pp. 1901-1921.

Payatakes, A.C., Ng, K.M., Flumerfelt, R.W., (1980). Oil ganglion dynamics during immiscible displacement: Model formulation. Am. Inst. Chem. Eng. J. 26, Pp. 430-443.

Payatakes, A.C., (1982). Dynamics of Oil Gauglia During Immiscible Displacement in Water-Wet Porous Media. Annual Review of Fluid Mechanics,Vol. 14, Pp. 365-393.

Rajagopalan, R. and C. Tien, (1976). Trajectory Analysis of Deep-Bed Filtration with the Sphere-in-cell Porous Media Model, Am. Inst. Chem. Eng. J. 22, Pp. 523-533.

Tobiason, J.E., and C. O'Melia, (1988). Physiochemial Aspects of Particle Removal in Depth Filtration. J. Am. Water Works Assoc. 80, (12). Pp. 54-64.

Wilson, J.L. and Conrad, S.H., (1984). Is Physical Displacement of Residual Hydrocarbons a Realistic Possibility in Aquifer Restoration? Proc. NWWA/API Conf. on Petroleum Hydrocarbons and Organic Chemicals in Ground Water - Prevention, Detection and Restoration, Pp. 274-298.

Yao, K.M., M.T. Habibian, and C.R. O'Melia, (1971). Water and Wastewater Filtration: Concepts and Applications. Evniron. Sci. Technol. 5, Pp. 1105-1112.