

Electro-Osmotic Remediation of Fine-Grained Sediments

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ELECTRO-OSMOTIC REMEDIATION OF FINE-GRAINED SEDIMENTS

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The coupled-flow phenomenon, electro-osmosis, whereby water flow results from an applied electrical potential gradient, is being used at Lawrence Livermore National Laboratory to induce water flow through deep (25-40 meters below surface) fine-grained sediments. The scoping work described here lays the groundwork for implementation of this technology to remediate solvent-contaminated clayey zones at the LLNL site. The electro-osmotic conductivity (k_e) measured *in-situ* between two 37 m deep wells, 3 m apart of $2.3 \times 10^{-9} \text{ m}^2/\text{s-V}$ is in good agreement with the value determined from bench-top studies on the core extracted from one of the wells of $0.94 \pm 0.29 \times 10^{-9} \text{ m}^2/\text{s-V}$. Hydraulic conductivity (k_h) of the same core is measured to be $2.03 \pm 0.36 \times 10^{-10} \text{ m/s}$. Thus, a voltage gradient of 1 V/cm produces an effective hydraulic conductivity of $\sim 1 \times 10^{-7} \text{ m/s}$; an increase in conductivity of nearly three orders of magnitude.

Despite on-going remediation efforts utilizing a variety of technologies, fine-grained sediments contaminated with organic solvents remain recalcitrant. These contaminated fine-grained areas are sources, slowly diffusing dissolved contaminants into adjacent high-permeability zones, leading to groundwater contamination. We are exploring the use of *in-situ* electro-osmotic pumping, with employment of this technology to flush contaminants from fine-grained sediments as our goal.

Electro-osmotic pumping is a known technology with applications in structural engineering (soil stabilization) (1), mining (sludge dewatering) (2), and remediation (soil cleanup) (3-6). Electro-osmotic soil remediation technology employs electrodes placed in the ground with a direct current (DC) passed between them using an external power supply. Clays have a net negative surface charge, balanced by loosely adsorbed (exchangeable) cations. Electro-osmosis is a secondary effect arising from electromigration of these cations through the porous matrix under an applied electrical potential (7). The flow of current results in movement of the cations and their associated water of hydration from anode to cathode, entraining contaminants, if present in the pores, in the flow. Electro-osmotic pumping can increase well yield in fine-grained sediments two to three orders of magnitude over flow rates achievable by hydraulic pumping alone. Contaminated water delivered to the cathode by electro-osmosis may then be mechanically pumped from the cathode well, and contaminants removed. The electro-osmotic conductivities of fine-grained clays with very low hydraulic conductivities (as low as 10^{-11} m/s) and of larger-grained sands with hydraulic conductivities of $\sim 10^{-6}$ m/s lie within the same narrow range, from 10^{-9} to 10^{-8} m²/s-V. Thus, for soils with very low hydraulic conductivities, rendering standard mechanical pump-and-treat technology virtually ineffective and costly, electro-osmotic pumping can greatly accelerate contaminant removal. Electrokinetic remediation has been shown useful for extraction of ions such as heavy metals, as well as organic chemicals from fine-grained soils (3-6).

The exploratory field work described here was conducted at the Lawrence Livermore National Laboratory site, in an area contaminated by fuel hydrocarbons from former underground storage tanks. This area has been the subject of extensive prior remediation and investigation efforts since the early 1990s (8). Cleanup technologies included dynamic underground steam stripping, electrical resistive heating, soil vapor extraction, groundwater extraction, and passive bioremediation. As a consequence of the subsurface investigations associated with these activities, the hydrostratigraphy in this area has been well characterized on a local scale. Of particular interest is a zone of predominantly fine-grained sediments located between 34 and 37 m below ground surface. Four electrode wells were installed in this area in a 3 x 3 m square arrangement, and the core from the screened portion of one of these wells preserved for use in bench-top cell measurements.

Bench-top cell measurements of electrical (σ_e), hydraulic (k_h) and electro-osmotic (k_e) conductivities of a soil core were performed in a pressure vessel, simulating actual underground pressures. Simultaneously, electro-osmotic pumping technology was deployed in the field where electrical and electro-osmotic conductivity were measured between electrode wells.

EXPERIMENTAL

We measured several parameters in both the bench-top cell and in the field. Electrical conductivity, σ_e , is determined using Ohm's Law, $\sigma_e = IL/EA$, where I is the current, L is the distance between electrodes, E is the voltage drop and A is the cross-sectional area of the core. The electro-osmotic conductivity, k_e , is calculated using $k_e = q_e L/EA$, where q_e is the electro-osmotic flux. Hydraulic conductivity, k_h , is obtained using Darcy's Law, $k_h = q_h L/HA$, where q_h is the hydraulic flux and H is the hydraulic pressure gradient. The power efficiency for electro-osmotic pumping, $P_{\text{eff}} = EI/q_e$ is also reported.

Bench-top Cell

We have designed and built a test cell to measure electro-osmotically induced flow, hydraulically induced flow, electric current and voltage distributions. *In-situ* conditions are simulated by subjecting the sample to a confining pressure matching the underground stresses of the original location of the soil core. The core used in the measurements reported here was extracted from the 36.4-36.6 m depth of a well drilled for the field installation. In this area, the water table lies at 29 m, and the stresses on the core can thus be estimated to lie in the 0.21-0.42 MPa range. Therefore, all bench-top measurements were acquired with confining pressure of 0.21 MPa.

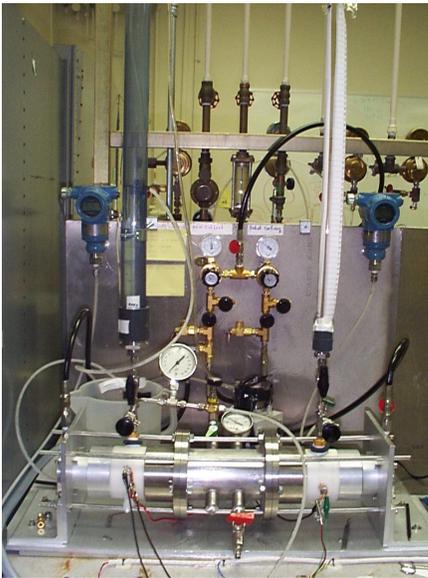


Figure 1a. Bench-top cell. The core is contained within the central pressure vessel, and water flow is measured as the level rises in the right hand (cathode) standpipe.



Figure 1b. Core assembly. The core is jacketed in Teflon shrinkwrap, with gold-plated copper perforated electrodes at either end, and two gold hoop voltage probes to provide information about the voltage drop along the core.

The test cell (Figure 1a) consists of a pressure vessel holding a 9 cm diameter by 15 cm long soil core (Figure 1b). The core is jacketed with a Teflon sleeve to seal against the confining pressure and to avoid short circuiting of the water flow at the circumference of the sample. Two perforated gold plated copper electrodes (anode and cathode) are placed on each end of the sample, and gold wire hoops placed around the core, 2 inches from each electrode, for use as voltage probes. The gold-plated diffusion plates are used to transfer the applied longitudinal load to the sample, as well as serving as electrodes. They are separated from the soil by a microporous membrane (Pall-RAI Electropore E40201 ultra high MW polyethylene, 100 μm thick, 2 μm pores). A 0-50 V Hewlett-Packard 6633B power supply was employed in DC constant voltage mode for electro-osmotic conductivity measurements. Water is supplied to the anode side of the cell by a constant hydraulic head standpipe during electro-osmotic flow measurements and by a pressurized water vessel for hydraulic flow measurements. A narrow diameter standpipe, outfitted with a 0-1.25 psi (0-8618 Pa) pressure transducer (Validyne DP 215-50), is used to measure water flow at the cathode side.

Field Installation

The equipment configuration in the field consisted of 3 m by 7.6 cm diameter graphite electrodes, installed within the screened intervals of two wells, W-1514 and W-1515 (15.2 cm diameter well bores). Two 3 m by 5 cm diameter carbon steel or graphite electrodes were installed in two other wells, W-1115 and W-1513 (4 inch diameter well bores). The four wells form a 3 x 3 m grid. Two 100 V/ 10 A power supplies provided power to the electrodes. Variable speed submersible pumps were placed in each of the wells, W-1514 and W-1515, housing the cathodes. The pumps in these two wells were plumbed to a set of instrumented manifolds. The treatment system for the extracted groundwater consisted of a series of granular activated carbon beds. Water was pumped from each cathode well at 3.8 L/min with a combined flow stream through the treatment unit at 7.6 L/min. After treatment the water was re-injected into the anode wells (W-1115 and W-1513). Sample ports were located at the influent and effluent streams and between the treatment units.

RESULTS AND DISCUSSION

Electrical Conductivity

In the bench-top cell, the voltage imposed between the anode and cathode (at either end of the 15 cm long soil core) is controlled in constant voltage mode. Further detail about the voltage drop along the core is provided by two supplemental gold hoop voltage probes at 5 and 10 cm along the core. The voltage difference between the anode and cathode is then V_{14} , and the voltage difference between the two passive voltage probes is V_{23} . Figure 2 shows the current-voltage plots of V_{14} and V_{23} to be linear, providing a soil electrical conductivity of 0.077 ± 0.020 S/m.

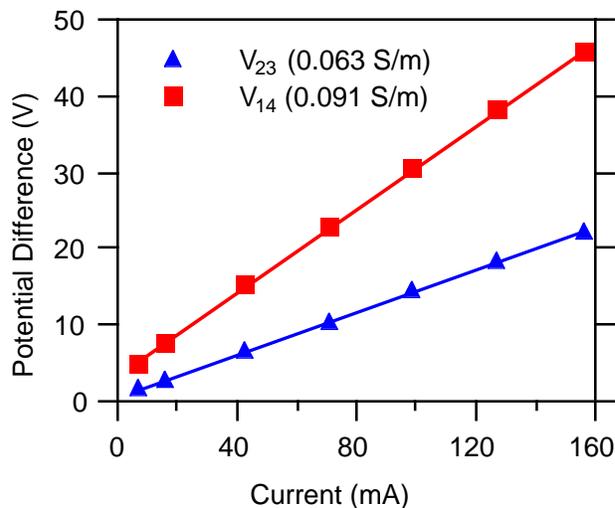


Figure 2. Electrical conductivity of the core is determined in the bench-top cell using Ohm's law. V_{14} is the current-voltage curve for the entire 15 cm core, V_{23} shows voltage drop between the two probes, each 5 cm from a working electrode and 5 cm apart, vs. current. The average electrical conductivity is measured at 0.077 ± 0.020 S/m.

Electrical resistivity in the field was measured using a four electrode array, with a voltage difference applied across the electrodes in wells W-1513 and W-1515 while the passive electrodes in W-1115 and W-1514 were used to monitor the voltage potential distribution. The advantage of measuring the voltage difference between the passive electrodes, as opposed to the active ones, is that voltage drops associated with surface chemistry effects, as well as those associated with the well water and the PVC well casing, could be avoided. With the chosen electrode geometry, the two passive voltage probes do not lie on an equipotential line, so a voltage difference can be measured.

The measured electric currents and passive electrode voltage differences in the field as a function of applied voltage across the active electrodes are shown in Table I.

Because the two passive electrodes consisted of unlike materials (carbon steel and graphite), a DC offset associated with their standard potentials of 0.683 V was subtracted from the data to yield the correct voltage difference. Based on the observed passive electrode voltage differences and the electrode geometry, a semi-analytical model of the potential distribution was used to calculate the soil bulk electrical conductivity (9). The resulting estimated value, approximately 0.13 S/m, is in good agreement with the bench-top measurement of ~0.08 S/m, and well within the typical range reported for soils, 0.01 to 1 S/m (7).

Table I. Four electrode soil conductivity test in the field.

Applied Voltage¹ (V)	Current (A)	Voltage Difference² (V)	Voltage Difference corrected³ (V)
10.20	0.9	0.792	0.109
20.03	2.1	0.918	0.235
30.02	3.4	1.035	0.352
40.00	4.6	1.158	0.475
50.03	5.6	1.281	0.598

¹Voltage applied to two graphite working electrodes in wells W-1513, W-1515

²Voltage difference between the two passive electrodes in wells W-1514, W-1115

³Voltage difference corrected for the 0.683 V zero current offset due to the standard potential difference between the carbon steel passive electrode in W-1115 and the graphite passive electrode in W-1514.

Hydraulic Conductivity

The standard technology used for remediation of organic solvent contamination at the LLNL site, “pump-and-treat”, is based on pumping water through contaminated zones, extracting contaminated water, and removing the contaminants. Hydraulic flow through heterogeneous lithologies preferentially passes through sandy, permeable zones, resulting in very little penetration of clayey, fine-grained zones. It is for this reason, that we are exploring electro-osmotic pumping to specifically address the finer-grained, less permeable sediments. The core chosen for work in the bench-top cell was selected due to its high clay content; representative of the finer-grained layers in the screened zone of one of the wells. Therefore, the hydraulic conductivity measured for this core is not directly relevant to the overall hydraulic permeability of the field installation, which contained several sandy layers within the screened zone, but rather is indicative of the type of sediments we are interested in targeting for cleanup with electro-osmotic pumping.

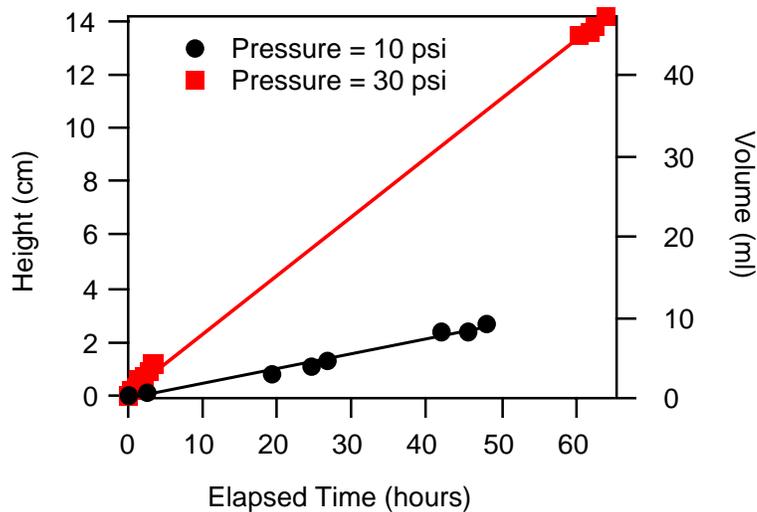


Figure 3. The hydraulic conductivity of the core was measured in the bench-top cell with 10 and 30 psi (0.069 and 0.21 MPa) hydraulic head gradients, providing an average calculated k_h of $2.03 \pm 0.36 \times 10^{-10}$ m/s.

Hydraulic conductivity was measured for the core in the bench-top cell, using a pressure differential imposed by a pressure can, pressurized with compressed air, on the inlet side and a standpipe open to atmospheric pressure at the outlet side. Flow rates using pressure gradients of 0.069 and 0.21 MPa were measured at 0.0030 and 0.012 ml/min, respectively (Figure 3). This corresponds to a hydraulic conductivity for this core of $k_h = 2.03 \pm 0.36 \times 10^{-10}$ m/s. Sediments with hydraulic conductivities in this range may be considered essentially impermeable to mechanical pumping, especially when interleaving sandy layers ($k_h > 10^{-6}$ m/s) are present.

Electro-osmotic Conductivity

Electro-osmotic conductivity (k_e) measurements may be performed under controlled conditions in the bench-top cell. Two measurements using the bench-top cell are presented in Figure 4. A 0.66 V/cm gradient results in a measured electro-osmotic flow rate or q_e of 0.028 ml/min, while 3 V/cm applied voltage resulted in $q_e = 0.082$ ml/min. The electro-osmotic conductivity for the 3.5 inch diameter core calculated from these measurements is $k_e = 0.94 \pm 0.29 \times 10^{-9}$ m²/s-V. However, the sediment sample used is small, isolated from the natural hydraulic gradients, and represents the finer-grained zones of the natural heterogeneous fabric. Therefore, in order to better understand the issues involved in field implementation of electro-osmotic pumping technology, we undertook not only bench-top measurements, but a field test based on use of a tracer that moves under electro-osmotic pumping. The most suitable tracer identified for this test proved to be water itself, labeled isotopically by its oxygen-18 (¹⁸O) fraction.

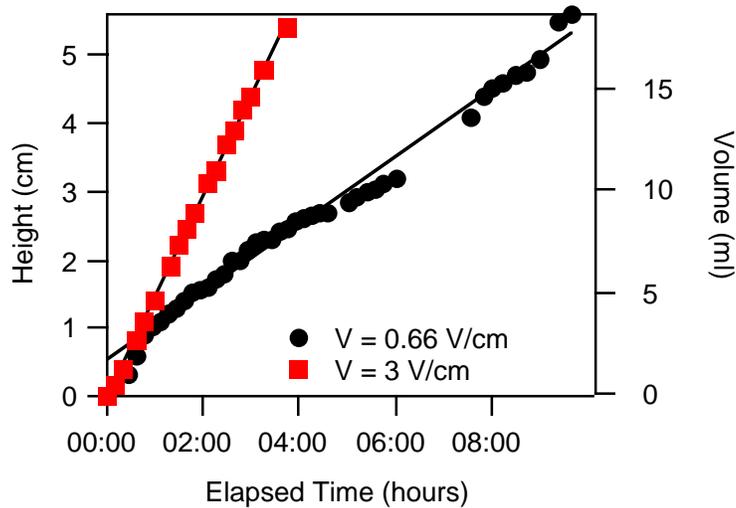


Figure 4. The electro-osmotic conductivity of the core was measured in the bench-top cell using 0.66 and 3 V/cm voltage gradients, providing an average calculated k_e of $0.94 \pm 0.29 \cdot 10^{-9} \text{ m}^2/\text{s}\cdot\text{V}$.

The tracer test employed for field measurement of k_e entailed placing tracer water (water with higher ^{18}O concentration than the natural groundwater) within a well bore which could be operated as either anode or cathode, and measuring the rate of loss of the tracer with the DC voltage on vs. the loss rate with the voltage off. The rate of loss of tracer above background loss is indicative of the electro-osmotic flux. A mechanical circulation system was devised to pump water from the well bore up to the surface and back into the well, allowing simultaneous injection of the tracer at the bottom of the well, at 3.8 L/min, while native groundwater was extracted at an equivalent rate from the top of the water level, thus minimizing differences in hydraulic head between the well bore and the surrounding aquifer.

Two tests were carried out using an applied voltage of 50 V between wells W-1514 and W-1515, first with W-1515 operating as an anode and second with the polarization reversed and well W-1515 as a cathode. For each test, a background test without applied voltage was run in sequence. Each test lasted for approximately 3 days. The well bore tracer fraction data indicate that approximately half of the tracer is lost from the well bore at the start of each of the tests. Much of this loss may result from density differences between the tracer water (25-30 °C) and native groundwater. Groundwater in the TFF area remains at elevated temperature (35 to 40 °C) several years after thermal treatment approaches (steam injection, electrical resistive heating) as a result of relatively slow rates of groundwater movement and the thermal insulating properties of the sediments. Presumably, the colder and thus denser tracer water, injected at the base of the water column, tended to flow out of the well screen into the surrounding sand pack as well as into

the sand fingers of the formation. When used as an anode, water will be generally drawn away from the vicinity of the well bore, but some of the tracer water which was initially lost to the surrounding sandy layers will be drawn back into the well, thus reducing the apparent rate of tracer loss in the well compared to the voltage-off background. When the well is used as a cathode, water will be drawn toward the well from the formation, moving tracer-laden water in the sand pack back into the well bore. Thus, with either electrode polarity, the tracer would be expected to remain longer within the well bore when a DC voltage is applied than when it is not.

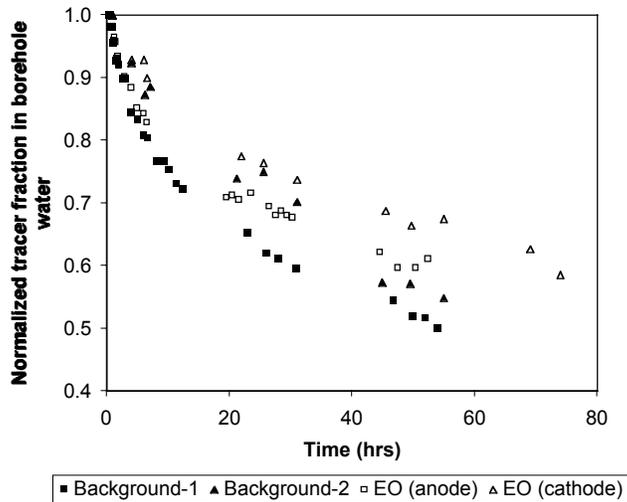


Figure 5. Tracer is lost in the well bore due to diffusion (Background, closed symbols), but loss of tracer is retarded when an electrical potential is imposed between electrode wells (EO, open symbols). Whether the well bore is an anode or a cathode, this is due to tracer water lost to the formation during injection being drawn back into the well by the electric field.

The tracer data from the two tests and background runs are shown in Figure 5, normalized to the tracer concentration at the beginning of the monitoring period. This indicates how much of the well bore water has mixed with native groundwater following the initial tracer loss at the start of the test. The changes in well bore water fraction over time reflect several mechanisms, including natural groundwater advection (focused into the well by the relatively high permeability of the sand pack), along with dispersive mixing across the well screen, sand pack, and surrounding formation (enhanced by the recirculating pumping action). Nevertheless, for measuring the electro-osmotic flux, it is only the *differences* between the curves for the cases when electric potential is applied, and when it is not, that are of interest. Therefore, for each of the two test pairs (first background test plus W-1515 as an anode, second background test plus W-1515 as a cathode), differences in well bore water fraction as a function of time were calculated. Specifically, for each sampling event from tests conducted with an applied voltage, the difference between well bore water fraction and the corresponding well bore water fraction without the applied current were calculated. Mismatches in sampling times between the tests were addressed using linear interpolation between the sampling events when the electric field was not present. These differences, as a function of time, are

shown in Figures 6a and 6b. For both tests, linear regression indicates a significant linear trend, with a slope of 0.002 %/hr, corresponding to approximately 6.1 L/day given the 125 L of water within the well bore. For comparison, a similar analysis of the differences between the two background runs indicates no relationship. This result suggests that differences between the respective background and DC voltage tests are not likely to be merely the result of chance.

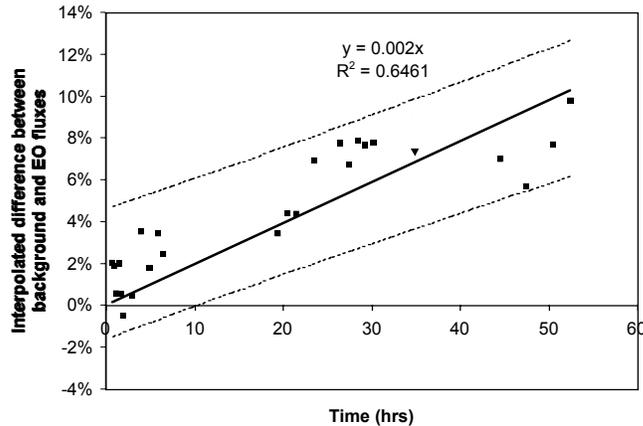


Figure 6a. The difference between background and background plus electro-osmotic flux is shown for the case where the well was operated as a cathode falls on a line corresponding to a flux of 6.1 L/day.

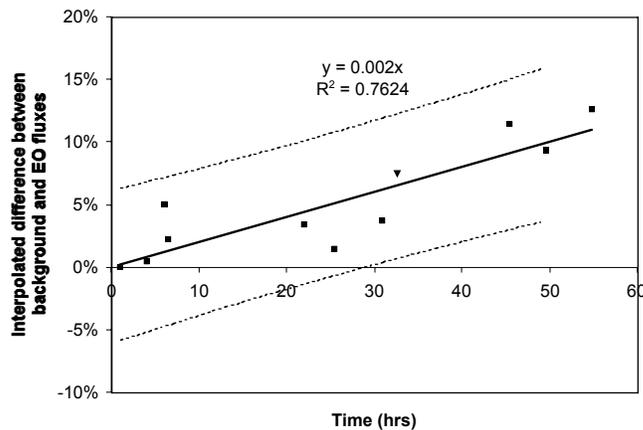


Figure 6b. The difference between background and background plus electro-osmotic flux is shown for the case where the well was operated as an anode falls on a line corresponding to a flux of ~6.1 L/day. These data and the data from Figure 6a were used in a model to determine the electro-osmotic conductivity at the field site.

The estimated electro-osmotic fluxes, taken with the electrode geometry, electric current, and bulk soil electrical conductivity can be used to estimate the bulk electro-osmotic conductivity. This estimation uses a semi-analytical model which utilizes a point

source solution to the steady-state potential field problem, integrated in the vertical direction to simulate a line source (i.e., an electrode), to predict the potential (i.e., voltage) as a function of position with respect to the line source. Superposition allows for multiple electrodes, with the sign on the current flow through each electrode used to distinguish anodes and cathodes. Thus, the model accepts current flow as input and calculates the voltage difference across the electrodes. The model is based on a number of simplifying assumptions, most notably that the soil electrical conductivity is homogeneous and constant in time and that the three-dimensional model domain extends infinitely in all directions. The model also assumes that the line sources do not depart significantly from a vertical orientation.

The construction of the semi-analytical model was performed using the MathCad (MathSoft, Inc.) computational environment. As the electro-osmotic velocity of water is, as an engineering approximation, proportional to the voltage gradient, the semi-analytical model used to calculate the voltage potential distribution can easily be extended to calculate the local electro-osmotic velocity field. That is, at any point in the model domain, the local groundwater velocity due strictly to electro-osmosis is given by $q_e = k_e \nabla\phi / n$, where the electro-osmotic conductivity, k_e , and the porosity, n , are input parameters and the voltage gradient ($\nabla\phi$) is calculated by numerical differentiation of the voltage potential line source model. Indeed, with a relationship in hand to quantify the flow field, the calculated summation of the electro-osmotic fluxes across a cylindrical surface surrounding an electrode will provide an approximation of the flux of water to the well (neglecting contributions across the top and the base of the cylinder). This calculation allows k_e to be adjusted in the model so that the predicted flux matches that observed in field tracer tests. The resulting estimate for k_e , roughly $2.3 \times 10^{-9} \text{ m}^2/\text{s-V}$, is in good agreement with the bench-top measurement of $\sim 1 \times 10^{-9} \text{ m}^2/\text{s-V}$, and within the typical range reported for soils, $1 - 10 \times 10^{-9} \text{ m}^2/\text{s-V}$ (7).

Power Efficiency

While electro-osmotic flux is directly proportional to the applied voltage, the cost of power is the product of the voltage and the current used. Electric power consumption over electro-osmotically pumped water flux is the power efficiency (Table II). The increased cost per voltage in the field reflects the substantial voltage drop across the PCV well casing, while the electrodes in the bench-top cell are separated from the core by only a thin microporous membrane with negligible resistivity. It is worthwhile to note (as demonstrated by the test cell measurements) that power costs will decrease for treatment at a low flow rate. This suggests that electro-osmotic pumping for cleanup of contaminated sediments may be considerably cheaper if treatment is carried out at lower voltages, over longer times.

Table II. Electro-osmotic power efficiency in the field and in the bench-top test cell.

Voltage (V/cm)	Average Current (A)	k_e ($m^2/s-V$)	$k_{h-eq.}$ (m/s)¹	P_{eff} (kWh/L)	Type of measurement
0.12	4	2.3×10^{-9}	2.8×10^{-8}	0.79	Field
0.66	0.0227	1.14×10^{-9}	7.5×10^{-8}	0.14	Test cell
3.00	0.1658	0.74×10^{-9}	2.2×10^{-7}	1.5	Test cell

¹Equivalent hydraulic conductivity, $k_{h-eq.}$, is k_e times the voltage over the distance between electrodes.

Conclusions

The results presented here show good agreement between measurements of electro-osmotic conductivity in a bench-top test cell, using a core extracted from one of the electrode wells drilled as part of the field installation, and measurements of water flux under electro-osmosis in the field wells. Systems for imposing a hydraulic gradient in a heterogeneous matrix will draw water primarily through the coarse-grained zones, rather than addressing the finer-grained zones where higher contamination is found. For this reason, standard mechanical pump-and-treat technology is not effective in cleanup of sediments such as that studied in the bench-top cell, with measured hydraulic conductivity of $\sim 2 \times 10^{-10}$ m/s. However, electro-osmotic pumping may be used to drive water through such sediments, cleaning up “impermeable” zones. From the electro-osmotic conductivity measured for the core in the bench-top experiments, $\sim 1 \times 10^{-9}$ $m^2/s-V$, an “equivalent hydraulic conductivity” ($k_{h-eq.}$) under electro-osmotic pumping can be determined. For the core studied here, an applied voltage of 1 V/cm yields a $k_{h-eq.}$ of $\sim 1 \times 10^{-7}$ m/s. This equivalent hydraulic conductivity results in a flow through the fine-grained sediments 500 times greater than without the applied field ($k_h \sim 2 \times 10^{-10}$ m/s)! Electro-osmotic pumping technology thus offers great potential to clean up fine-grained sediments, even, as shown here, deeply buried (~ 40 m) contaminated zones. Advantages include: (1) Flow is controlled by location of electrode wells and applied potential polarity, and volume treated may be calculated based on the domain of the imposed electric field, (2) Fine-grained sediments may be specifically targeted due to their greater electrical conductivities, effectively channeling electro-osmotic flow through them, and (3) Electro-osmosis is an *in-situ* cleanup technology which requires no excavation, nor significant chemical by-product residue. The next phase of our work will explore removal efficiency of organic solvents from contaminated sediments under electro-osmotic pumping both in the field and in the bench-top test cell.

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KEYWORDS

soil remediation, electrokinetics, electro-osmosis, electromigration, electroremediation,
flow in porous media