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Lead (II) Removal from Natural Sandy Soils by Enhanced Electrokinetic Remediation

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Electrokinetics is the movement of ions and charged solid particles between two electrodes under the influence of an electrical field. This phenomenon can be used many fields related to civil and environmental engineering (such as consolidation and stabilization of soils, dewatering in construction sites, decontamination of pollutants (called as electrokinetic remediation), injection of chemical agents to soil etc.) (Chung and Kang 1999).

In recent years, a number of laboratory-scale and field scale studies have shown that electrokinetic remediation can be effectively used to metal removal from soils (Yang and Lin 1998). When DC electric fields are applied to contaminated soil, migration of charged ions occurs. This migration is toward the anode for anionic species and toward the cathode for cationic species (Virkutyte et al., 2002). Previous researches have shown that the principal of contaminant migration mechanisms that occur during the electrokinetic remediation process are electroosmosis, electromigration, diffusion and electrophoresis (Acar and Alshawabkeh 1993; Probstein and Hicks 1993; Reddy et al. 1997). But, contaminant transport is generally governed by electroosmosis and electromigration (Puppala et al. 1997). Electroosmosis is the movement of pore water under electrical potential difference from the anode to cathode. Contaminant transport occurs by this movement. Electromigration is the movement of ions in the pore water of the soil under the influence of an electric current (Reddy et al. 1997).

Numerous electro chemical reactions and soil contaminant interactions such as electrolyses, sorption-desorption and precipitation occur simultaneously during electrokinetic remediation (Chung and Kang 1999). Some of these reactions affect to electrokinetic remediation negatively. For example, electrolyses reactions at electrodes generate acidic medium (pH 2-3) at anode and alkaline medium (pH 10-12) at cathode (Reddy and Shirani 1997). In this situation, the low pH values near the anode promotes desorption of heavy metals and other cations. Conversely, the high pH values near the cathode results in the precipitation of cations (Hicks and Tondorf 1994). These precipitation reactions can be hindered by different enhancement techniques such as using purging solutions (AcH, citric

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acid, HCl etc.) or complexing agents (EDTA etc.) and special electrodes and membranes (Puppala et al. 1997; Yang and Lin 1998).

The success of electrokinetic remediation depends on mineralogical structure of soil. Previous studies have shown that electrokinetic remediation process is applied effectively to clays with low sorption and buffering capacity (Hamed et al. 1991). However, the studies related to natural soils except for glacial till, marine clay, river slush, silt-sand mix, fine sand and silty loam etc. are rather limited (Reddy and Shirani 1997; Chung and Kang 1999; Lageman 1993; Yang and Lin 1998).

In this study, the feasibility of using enhanced electrokinetic remediation to remove lead (II) from sandy and silt sandy soils were investigated under different controlled laboratory conditions.

MATERIALS AND METHODS

In this study, different two soil sample used. First sample (silty sandy soil) was taken from near the Kumarli Landfill area and second sample (sandy soil) was taken from near the zinc production plant in Kayseri City (Turkey). The properties of these samples and using experimental methods are summarized in Table 1.

Electrokinetic remediation apparatus adapted from Hsu (1997) is primarily consisted of an electrokinetic extraction cell, fluid volume and gas volume measurement devices, a DC electrokinetic power supply and a multimeter (Figure 1). Electrokinetic extraction cell includes a specimen cylinder (7.5*15 mm, D*L) made of PVC, two end flanges that house a purging solution reservoir and graphite electrodes (7.5*3 mm, D*W) and voltage measurement probes made of steel. The flow volume and gas volume measurement devices are consisted of four 350 mm long glass cylinders. Internal diameter of cylinders used gas measurement and fluid measurement is 12 mm and 35 mm, respectively.

For silty sandy soil, calculated concentrations of $Pb(NO_3)$ solution were added to 1.5 kg soil sample, in which the soils lead concentration was converted to about 1000 mg/kg and 4000 mg/kg. The soil contaminant mixture was manually mixed. Then, it was allowed to equilibrate for 48 hours. Prepared slurry was transferred into specimen cylinder and compacted by consolidation method of described in Reddy et al. (1997). Finally, all excess found at edges of the cell was shaved away and specimen cylinder was assembled in electrokinetic extraction cell.

In this study, silty sandy soil sample was contaminated by lead (II), in which soil lead concentrations were converted to about 1000 mg/kg and 4000 mg/kg. After soil samples assembled in electrokinetic apparatus, electrokinetic remediation process applied to these samples at 10-20 volt constant potential and different test conditions. Testing program is summarized in Table 2. Test conditions of sandy soil contaminated with a zinc production wastes was determined according to results obtained from silty sandy soil tests.

Table 1. General properties of soil samples.

Parameter	Silty Sandy Soil	Sandy Soil	Method
Grain size distribution			ASTM (1995a) and ASTM (1995b)
Coarse sand	%8	%10	
Medium sand	%50	%52	
Fine sand	%21	%30	
Silt	%17	%6	
Clay	%4	%2	X-Ray Diffraction
Mineralogy			
Quartz	%36	%45	
Feldspar	%33	%47	
Gypsum	%1	---	
Calcite	%2	---	
Clinoptilolite	%24	%6	
Smectite	%3	%2	
Illite	%1	---	
Soil classification	Silty sand	Sand	USBR (1974)
Pb (II) exchange cap.	8.8	4.7	Reddy et al. (1997)
Porosity	0.36	0.32	Calculation

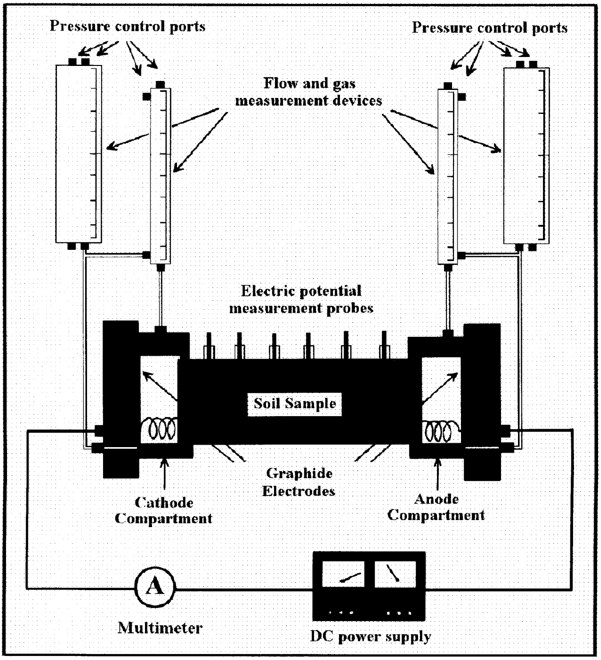


Figure 1. Electrokinetic remediation apparatus (adapted from Hsu (1997)).

Table 2. Summary of testing program.

Sample	Test No	Initial Pb (ppm)	Voltage (volt)	Test Duration (hour)	pH	Purging Solution
Silty Sand	Test-1	4000	20	233	6.85	H_2O
	Test-2	4000	10	137	7.40	3.0 M AcOH
	Test-3	4000	10	473	7.50	0.5 M AcOH
	Test-4	1000	20	220	7.53	3.0 M AcOH
	Test-5	1000	10	240	7.30	3.0 M AcOH
Sand	Test-6	2590	20	272	7.81	3.0 M AcOH
	Test-7	2610	20	272	7.78	0.5 M AcOH
	Test-8	2720	10	272	7.65	3.0 M AcOH
	Test-9	2676	10	272	7.60	0.5 M AcOH

During experimental studies, variations of current through the soil and flow-gas devices were observed depend on time. At the end of electrokinetic remediation tests, soil samples were disassembled from the apparatus and sliced into six sections. The values of pH and lead (II) concentration were measured in each slice. In the measurement of lead (II) and pH, methods were used suggest from Hamed et al. (1991) and McLean (1982).

RESULTS AND DISCUSSION

In electrokinetic remediation, electroosmosis, electromigration and diffusion are dominant driving forces (Acar and Alshawabkeh 1993). For this reason, electroosmotic flows in different test conditions were measured during experimental studies and results were given in Figure 2.

As seen in Figure 2, direction of electroosmotic flow in all tests is generally toward cathode from anode. Electroosmotic flow is directly related to soil zeta potential, which is a complex function of soil properties and chemical composition of pore water (Reddy et al. 2001). Increasing negativity of soil zeta potential, electroosmotic flow also increases. According to this, it may be said that zeta potential of silty sandy soil is more negative than sandy soil. Another result that is seen in Figure 2, electroosmotic flow increases by applying high voltage to system. It is shown clearly at Test-4, Test-5, Test-6, Test-7 and Test-9. Similar results were also reported by Shapiro and Probstein (1993). Enhanced electrokinetic remediation tests in this study were demonstrated that using AcH as purging solution in cathode reservoir affects to electroosmotic flow positively. This phenomenon relates with decrease of precipitation reactions in cathode region using AcH (Chung and Kang 1999). Similarly, Reddy et al. (2001) also pointed out that electroosmotic flow might be decreased by precipitation reactions at cathode region. When an electrical charge is applied to the saturated soils, an electrolysis reaction of water takes places at the electrodes. Oxygen and hydrogen ions are produced at the anode and hydrogen and hydroxide ions are produced at the cathode.

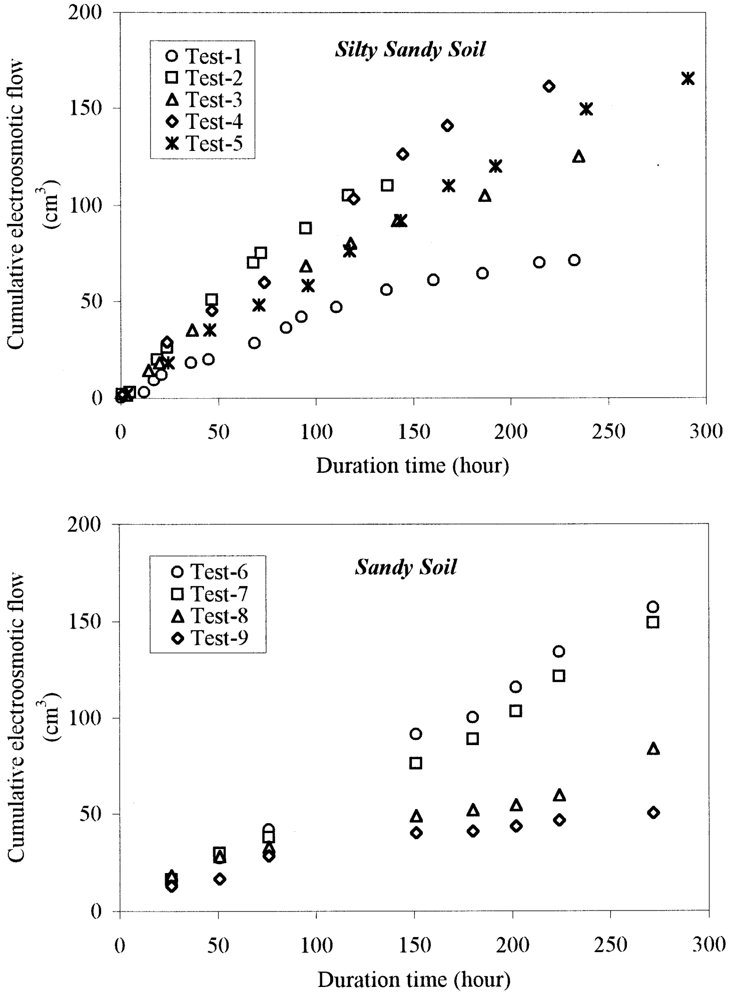
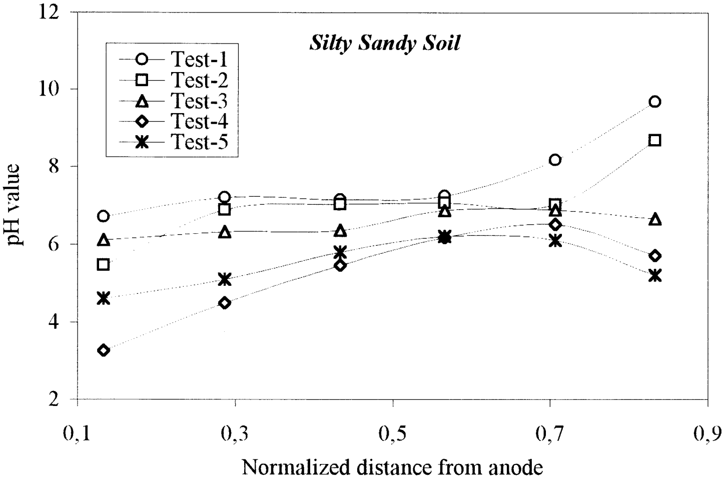


Figure 2. Variation of the electroosmotic flow with time under different test conditions.

These ions migrate toward cathode or anode depend on theirs polarities by electroosmosis, electromigration and diffusion (Acar et al. 1994). Thus, changing of soil pH profiles between anode and cathode hinders contaminant migration. For determination of these phenomena, pH variations of soil sample were measured and results obtained from these measurements were given in Figure 3. Puppala et al. (1997) determined that pH value less than 6 would be sufficient to dissolve and remove the lead (II) in the soil and aid in lead (II) transport to cathode. Virkutyte et al. (2002) also reported that precipitation reactions are minimum at pH value less than 4.5.



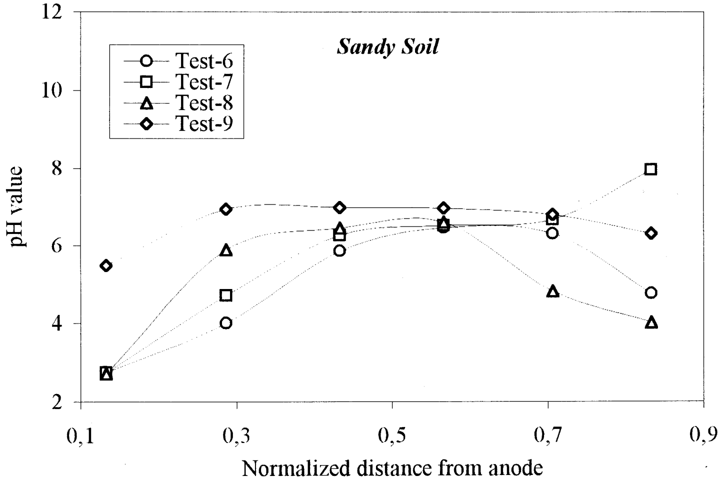


Figure 3. The pH profiles in electrokinetic remediation under different test conditions.

As seen in Figure 3, pH values at regions of closed to cathode increase up to 10 in unenhanced electrokinetic remediation tests. However, in all enhanced tests (except Test-7) these high pH values may be neutralized by using a purging solution involved 3.0 M and 0.5 molar AcH. In a study that was done by Yang and Lin (1998), this concentration for silty loam was determined as 0.1 M.

Migration velocity of H^+ ions towards cathode is very important on removal efficiency of metals by electrokinetic remediation. In this case, drag forces providing acid front migration are electroosmosis and electromigration. The most important parameter affecting to these forces is high of voltage applying to system. This case is clearly seen at results obtained from Test-4, Test-5, Test-6 and Test-8. As seen in Figure 3, acid front is reached to normalized distance between 0.3 and 0.4 at 220 or 272 hours in Test-4, Test-5 and Test-6. Similarly, Vengris et al. (2002) also were reported that this distance for sandy soil was 0.5 after 225 hour.

Heterogeneous soils may contain a lot of minerals with alkali and high sorption capacity such as calcite, dolomite, clinoptilolite. Electrokinetic remediation of these type soils is very difficult. As seen in Table 1, above mentioned minerals are present small quantities in soil samples using this study. For this reason, it is said that electrokinetic remediation of these samples is quite easy. However, electrokinetic remediation, which is a new technology, has mostly been applied to pure clays type and their synthetically prepared mixtures. For this reason, there are important knowledge deficiencies about applications of this technology to different soil types. Therefore, for contribute to solving of the above mentioned problems, the lead (II) removal efficiencies from sandy and silty sandy soils at different test conditions were determined and results were presented in Figure 4.

As seen in Figure 4, best results for sandy and silty sandy soils were obtained at adding 3.0 molar AcH and applying 20-volt constant potential (Test-4, and Test-6). In these test conditions, lead (II) removal efficiencies for sand soil and silty sandy soil were varied between 60%-80% and 65%-85% up to 0.6 normalized distances, respectively. Similarly, Sah and Chen (1998) were pointed out that this efficiency for sandy soil was 70% up to 0.7 normalized distances from anode at the end of seventh day. However, 0.1 M HCl as purging solution was used in that study. In other study for these type soils, Vengris et al. (2001) also obtained 70% lead (II) removal up to 0.5 normalized distance after 220th hours.

Another result that may be derivate from experimental studies is that lead (II) removal efficiencies of silty sandy soil sample were lower than sandy soil sample relatively. It is though that this difference in removal efficiency is occurred by clinoptilolite in silty sandy soil sample. As is know, because clinoptilolite minerals have high sorption capacity, contaminant migration in soils having these type minerals is rather limited.

Another important result shown in Figure 4 is that removal efficiency of electrokinetic remediation test at applying 20-volt constant potential is higher than others. Particularly, this difference is fairly shown at results obtained from Test-4, Test-5, Test-6 and Test-8. But, it is also taken into consideration that energy expenditure of system will increase proportionally at applying high voltage (Azzam and Oey 2001).

According to Chung and Kang (1998); the transport of lead (II) at low levels of concentration is slower, because almost all the lead is adsorbed to soil particle. But at high levels of concentration, the transport of lead (II) is faster, because most of the lead (II) is free in pore and not adsorbed to the soil particle.

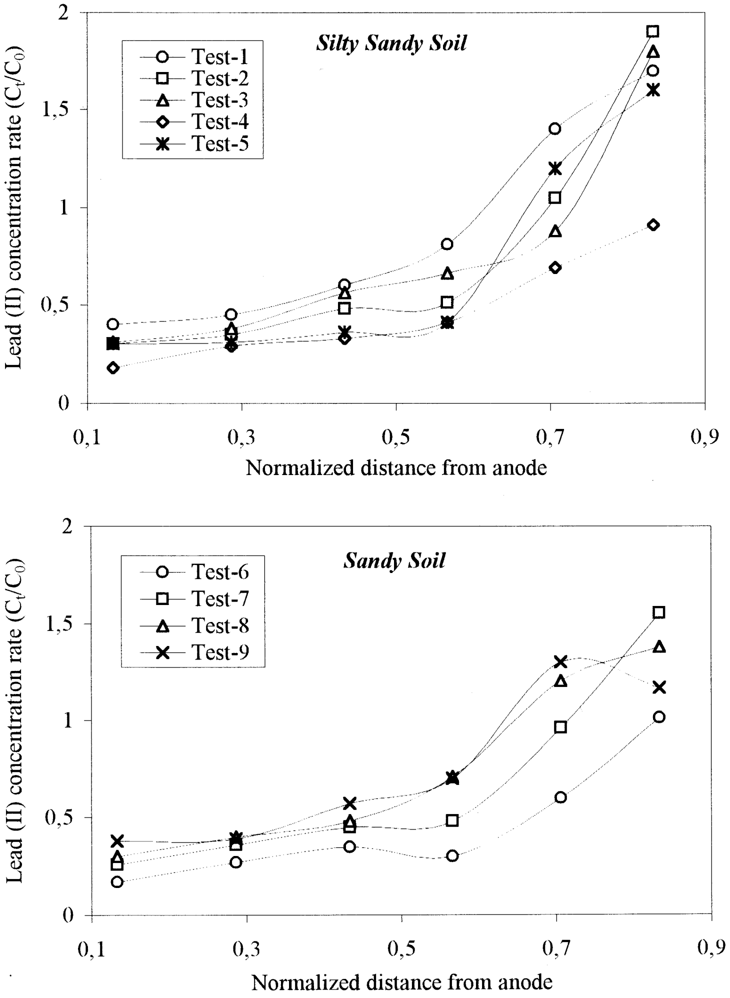


Figure 4. The Lead (II) profiles in electrokinetic remediation under different test conditions.

When results obtained from Test-2 are compared to Test-5, it is clearly shown that similar removal efficiencies for high contaminant concentration may be achieved short treatment time.

Conclusions derived from experimental studies have been summarized as follows:

- Enhanced electrokinetic remediation can be used effectively in sandy and silty sandy soils.
- The precipitation reactions occurring around cathode area may be hindered by using 0.5 M AcH. Nevertheless, lead (II) removal efficiencies obtained from

tests of using 3.0 M AcH were better. In addition to this, treatment time may be decreased by using 3.0 M AcH as a purging solution.

- Dominant forces behind the lead (II) transport during electrokinetic remediation of sandy and silty sandy soils are electromigration and electroosmosis in the initial hours. However, electromigration is higher than electroosmossis forward remediation time.

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