Effects of Soil Moisture and Heavy Metal Concentrations on Electrokinetic Remediation

Krishna R. Reddy*, Richard E. Saichek[†], Kranti Maturi[‡] and Prasanth Ala[‡]

Introduction

Accchording to the United States Environmental Protection Agency (USEPA), there are over 217,000 sites in the U.S. that urgently require remediation, and the estimated cost is over $187 billion U.S. dollars. The contaminants encountered include heavy metals, such as lead, chromium, strontium, and uranium, as well as volatile organic compounds, such as benzene, toluene, and trichloroethylene (TCE). There are also semi-volatile organic compounds, such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs). In addition, the metal contaminants and the volatile and/or semi-volatile organic contaminants frequently co-exist at these sites.

There are many remediation technologies available for treating contaminated soils and groundwater that are classified as either ex-situ or in-situ techniques (Reddy et al., 1999). Ex-situ techniques treat the contaminated soil and/or groundwater after it has been removed from the subsurface, whereas in-situ techniques treat the soil and/or groundwater within the subsurface itself. In-situ techniques are preferred because, generally, they cause less site disturbance, less contaminant exposure to the environmental professionals and public in the vicinity, and they are often less complicated and more economical. The in-situ technologies that are commonly used for remediating soils include soil vapor extraction (SVE), soil flushing,

* Associate Professor, University of Illinois at Chicago, Department of Civil and Materials Engineering, 842 West Taylor Street, Chicago, Illinois 60607, USA. E-mail: kreddy@uic.edu

[†] Research Associate
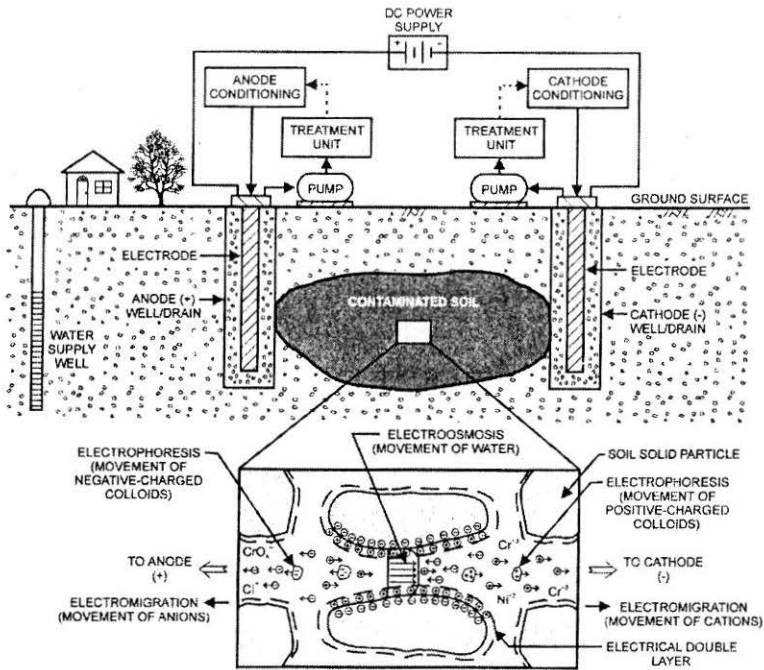
[‡] Graduate Research Assistant

FIGURE 1 : Schematic In-Situ Electrokinetic Remediation System

solidification/stabilization, chemical oxidation, soil heating, vitrification, bioremediation, electrokinetics, and phytoremediation. The common in-situ techniques for remediating groundwater include pump-and-treat, air/ozone sparging, flushing, permeable reactive barriers, immobilization, chemical oxidation, electrokinetics, and bioremediation.

Although many different in-situ soil remediation technologies are available, electrokinetics offers many advantages. This paper provides an overview of electrokinetic remediation, including the research performed at the University of Illinois at Chicago (UIC), and presents the new research to evaluate the effects of soil moisture content and contaminant concentration on the electrokinetic remediation process for heavy metal-contaminated soils.

Background

Research on electrokinetically induced transport mechanisms, particularly on electroosmosis, has been conducted for nearly two centuries (Casagrande, 1949; Yeung, 1994). Within the last few decades, however, environmental professionals began using the electrokinetic method for the remediation of contaminated soils and/or groundwater, and this technique has

been called electrokinetic remediation, electroremediation, electrorestoration, electroreclamation, electrochemical decontamination, or electromigration. A typical in-situ electrokinetic remediation system is shown schematically in Fig.1. Essentially, electrokinetics involves the installation of electrodes into the subsurface surrounding the contaminated region. After the electrodes are in place, a low electrical potential is applied across the anode(s) (positively charged electrode) and the cathode(s) (negatively charged electrode). As a result of the electrical gradient, different physico-chemical reactions occur and contaminant transport occurs due to various mechanisms within the soil and groundwater. Generally, for the migration to be significant, the contaminants should be in a soluble form. If they are not soluble, they need to be desorbed, dissolved, and/or solubilized into the pore solution before they can be adequately transported from the soil to an electrode wells/ reservoirs. During the process, or after the process is complete, the contaminant-laden solution is removed from the electrode wells/reservoirs, and the contaminants are subsequently extracted from the solution using conventional wastewater treatment methods. Depending on the type of contaminant and type of electrode, some contaminant adsorption may occur to the electrodes. Hence, the electrodes also require proper care and disposal. Pumping and/or optimal conditioning (or purging) solutions may be introduced for specific soil or contaminant conditions. Electrokinetics can also be used as an ex-situ treatment technology by employing an engineered reactor similar to the one shown schematically in Fig.2.

The successful implementation of the electrokinetic technique requires a thorough understanding of the physico-chemical reactions and the contaminant transport processes under an induced electric potential. Initially,
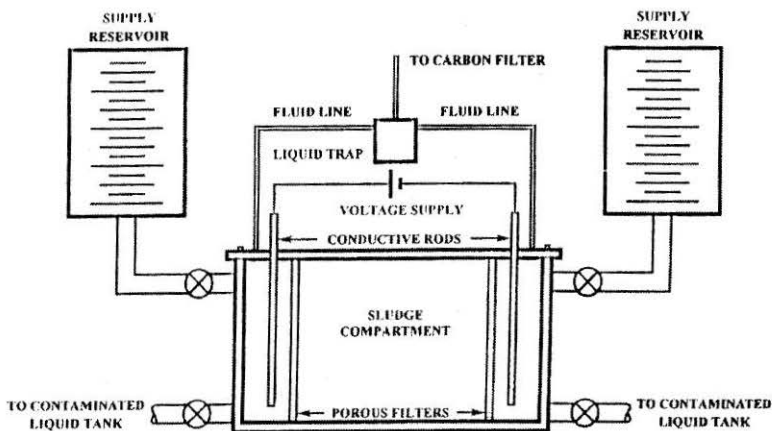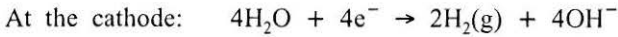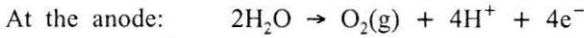

FIGURE 2 : Schematic Ex-Situ Electrokinetic Remediation System

it should be recognized that under an electric potential, the electrolysis of water occurs at the electrodes according to the following reactions:

At the anode: $2H_2O \rightarrow O_2(g) + 4H^+ + 4e^-$

At the cathode: $4H_2O + 4e^- \rightarrow 2H_2(g) + 4OH^-$

Thus, the electrolysis reactions cause an acidic solution to be generated at the anode and an alkaline solution at the cathode. The three major contaminant transport mechanisms in electrokinetic remediation are *electromigration*, *electroosmosis*, and *electrophoresis*. *Electromigration* refers to the transport of ionic species, such as the metal ions that are present in the pore fluid. This mechanism is largely responsible for generating the electrical current, and it includes the electromigration of H^+ and OH^- towards the oppositely charged electrode(s). Moreover, under an electric potential, *electroosmotic flow* is produced because the locally existing excess ions migrate in a plane parallel to the particle surface towards the oppositely charged electrode, and, as they migrate, they transfer momentum to the surrounding fluid molecules via viscous forces (Eykholt, 1992). Electroosmosis depends on the net amount of ionic migration towards an electrode location, and, in low permeability clayey soils, the net ion migration may be high and it could significantly affect the decontamination process. The following Helmholz–Smoluchowski (H-S) equation is often used to estimate the average electroosmotic flow velocity (v_{eo}) (Eykholt and Daniel, 1994):

$$v_{eo} = -\frac{D\varepsilon_0\zeta}{\eta}E_x$$

According to this equation, the flow velocity is proportional to the electrical gradient (E_x), zeta potential (ζ), and dielectric constant (D), and it is inversely proportional to the viscosity (η). The ε_0 term represents the permittivity of vacuum (8.854×10^{-12} C/V·m), and the dielectric constant and viscosity are properties of the pore fluid. The zeta potential depends on the zero point of charge (ZPC), which refers to the pH at which the net charge on the soil particle surface is zero. Generally, when the pH is below its ZPC, the soil particle surfaces have a positive zeta potential and the electroosmotic flow occurs towards the anode. Conversely, when the pH is above the ZPC, the soil particles have a negative zeta potential and the electroosmotic flow occurs towards the cathode. The electroosmotic flow velocity may also be expressed as:

$$v_{eo} = K_e E_x$$

where K_e is referred to as the electroosmotic conductivity. *Electrophoresis* is the migration of charged colloids, but, in a compact clay soil system, electrophoresis is less important since colloid-sized particles are generally restrained from movement.

The electrolysis reactions greatly affect the remediation process because the ionic products (H^+ and OH^-) may electromigrate and/or be transported by electroosmotic advection towards the oppositely charged electrode location. Thus, an acidic (H^+) front of solution may move from the anode towards the cathode, and/or an alkaline (OH^-) front of solution may move from the cathode towards the anode. The rate of electromigration may also be affected by ionic mobility, and, since hydrogen ions are smaller and have a greater mobility than hydroxyl ions, the acidic front generally moves faster through the soil (Acar and Alshawabkeh, 1993). The reaction kinetics, or the rate of the electrolysis reactions at the electrodes, may also affect the generation and movement of the hydrogen and/or hydroxyl ions.

From the H-S equation, it can be seen that changes in the surface charge of the soil particles (zeta potential) and changes in the pore fluid properties (such as dielectric constant and viscosity) influence the electroosmotic flow. In addition, the electrical gradient may not be uniform through the soil and hence the electroosmotic flow is generally not uniform spatially or temporally (Eykholt and Daniel, 1994). Therefore, the average electroosmotic conductivity (K_e) through the soil usually varies with time, and, as a result of these physico-chemical changes, the electroosmotic flow may cease or even reverse in direction.

Compared to conventional remediation technologies, electrokinetics has several advantages, such as the following:

• Simplicity: It is easy to operate and requires simple equipment.

• Safety: There is minimal exposure to the operating personnel and the surrounding environment.

• Economical: Costs range from \$20 to \$225 per cubic yard (depends on site specific conditions).

• Flexible: It can be used as an in-situ or ex-situ remediation system, as a delivery system, as a containment system, or as a combination of these systems.

• Applicable for a wide range of media: It may be used for soils, sludges, sediments, and/or groundwater. Electrokinetics is ideal for subsurface conditions that possess low permeability soils or soil deposits with

stratified layers or lenses of variable permeability. Here the conventional remedial methods are commonly deficient.

- Applicable for a variety of contaminants: It may be used for metals, volatile or semi-volatile organic compounds, and/or radionuclides, alone or combined.

Numerous laboratory bench-scale electrokinetic studies have been performed on a diverse range of contaminants. Many of these studies were conducted using kaolin soil and an individual heavy metal contaminant (Eykholt and Daniel, 1994; Hamed et al., 1991). In addition, electrokinetic remediation has been used at several contaminated sites, but the number of research studies has been limited and not well documented. Most of the field cleanup projects were for the removal of heavy metal contaminants (Lageman, 1993), but a recent large-scale project was for the remediation of a TCE-contaminated soil using the Lasagna process (Ho et al., 1999a,b). The Lasagna process is essentially an electrokinetic remediation method, where the electrodes and treatment zones are distributed across the site in a layered configuration. The results from this recent field investigation were encouraging, and an average TCE removal of 98% was achieved (Ho et al., 1999a).

Table 1 shows the summary of electrokinetic remediation projects completed at UIC. These studies have focused on the physico-chemical changes that occur within the soil during the electrokinetic treatment process, especially under different soil environments and with different types of spiked contaminants. The soil environment and/or composition, such as mineralogy, concentration of oxidizing or reducing species, and the carbonate and organic content, were found to significantly affect the electrokinetic remediation process. In addition, different types of contaminants have been investigated, either alone or in combination (synergistic effects), and research has been conducted to optimize the electrokinetic variables. The electrokinetic variables include the introduction of pre-selected purging solutions, the effect of increasing the voltage gradient, and/or the use of various remediation times.

Scope of the Present Study

In the previous UIC investigations that employed kaolin, a specific amount of water, or water-contaminant solution, was added to the soil so that the initial moisture content would be approximately 30%. This initial moisture content was used to simulate normal field conditions and to facilitate soil placement into the electrokinetic reactor. It was hypothesized that increasing the initial moisture content may assist contaminant migration and removal. Increasing the moisture content should theoretically increase the amount of solubilized contaminant as well as the amount of particle dispersion. This is

TABLE 1 : Electrokinetic Research Conducted at UIC

Investigators	Purpose	Soil(s)	Contaminant(s)	Purging Solution		Electric Potential [VDC/cm]
				Anode	Cathode	
Saichek (2002)	To evaluate the use of different surfactants/cosolvents for the electrokinetic removal of phenanthrene from kaolin	Kaolin	Phenanthrene (Polycyclic Aromatic Hydrocarbon) (PAH)	Deionized Water	Deionized Water	1.0
				1% Witconol-2722	Deionized Water	1.0
				3% Tween 80	Deionized Water	1.0
				40% Ethanol	Deionized Water	1.0
				4% Witco 207 and 40% ethanol	Deionized Water	1.0
Saichek (2002)	To investigate the effect of soil type for the electrokinetic removal of phenanthrene using surfactants or cosolvents	Kaolin and Glacial till	Phenanthrene (Polycyclic Aromatic Hydrocarbon) (PAH)	Deionized Water	Deionized Water	1.0
				Ethanol	Deionized Water	1.0
				Tween 80	Deionized Water	1.0
Saichek (2002)	To investigate the advantages of adding a buffer to the anode purging solution and surfactant or cosolvent in order to electrokinetically remove phenanthrene from kaolin	Kaolin	Phenanthrene (Polycyclic Aromatic Hydrocarbon) (PAH)	Deionized Water with and without NaOH	Deionized Water	1.0
				Ethanol with and without NaOH	Deionized Water	1.0
				Tween 80 with & without NaOH	Deionized Water	1.0

Saichek (2002)	To optimize the electrokinetic removal of phenanthrene from kaolin by using different buffering and surfactant concentrations and voltage gradients	Kaolin	Phenanthrene (Polycyclic Aromatic Hydrocarbon) (PAH)	Igepal CA-720 and NaOH buffer	Deionized Water	1.0, 2.0
Saichek (2002)	To investigate the use of a periodic electric potential application for the removal of phenanthrene from kaolin	Kaolin	Phenanthrene (Polycyclic Aromatic Hydrocarbon) (PAH)	Igepal CA-720 and NaOH buffer	Deionized Water	1.0, 2.0 (Periodic)
Reddy and Chinthamreddy (2002)	To evaluate the effects of the initial form of chromium on its remedial efficiency from different contaminated clays	Kaolin and Glacial till	Cr(VI), Cr(III), and Cr(VI) and Cr(III) combined	Potable Water	Potable Water	1.0
Reddy et al. (2001a)	To evaluate the effects of Cr, Ni and Cd when combined together compared to when these contaminants existed by themselves	Kaolin and Glacial till	Cr(VI), Ni(II), and Cd(II)	Potable Water	Potable Water	1.0
Reddy et al. (2001b)	To assess contaminant speciation and migration using a sequential extraction analysis procedure on the soil that was treated using electrokinetics	Kaolin and Glacial till	Cr(VI), Cr(III), Ni(II), and Cd(II)	Potable Water	Potable Water	1.0
Li et al. (2000)	To investigate the removal of phenanthrene from glacial till when different cosolvent purging solutions were used	Glacial till	Phenanthrene (Polycyclic Aromatic Hydrocarbon) (PAH)	Deionised Water	Deionised Water	1.0
				n-Butylamine	Deionized Water	1.0
				Acetone	Deionized Water	1.0
				Tetrahydrofuran	Deionized Water	1.0

Chaparro (1999)	To investigate the use of electrokinetics for Hg-contaminated soils	Kaolin and Glacial till	Hg(II)	Deionized Water	Na-EDTA, KI	1.0
Chaparro (1999)	To optimize the removal of Hg using KI solution	Kaolin and Glacial till	Hg(II)	Deionized Water	KI	1.0, 1.5
Chinthamreddy (1999)	To evaluate different purging solutions for the removal of multiple heavy metal contaminants from kaolin	Kaolin	Cr(VI), Ni(II), and Cd(II)	Potable Water	Potable Water	1.0
				Potable Water	Potable Water	1.0
				Potable Water	Acetic Acid	1.0
				Potable Water	EDTA	1.0
				NaOH	Acetic Acid	1.0
Chinthamreddy (1999)	To evaluate different purging solutions for the removal of multiple heavy metal contaminants from glacial till	Glacial till	Cr(VI), Ni(II), and Cd(II)	Potable Water	Potable Water	1.0
				Potable Water	EDTA	1.0
				Potable Water	Acetic Acid	1.0
				Potable Water	Citric Acid	1.0
				NaCl	EDTA	1.0
				Sulfuric Acid	Sulfuric Acid	1.0
Reddy et al. (1999a)	To assess electrokinetic performance on a site-specific field soil containing a mixed waste and sludge	Field soil and sludge	Metals (Cd, Cr(VI), Ca, Pb, Mg), VOCs, and Radionuclides	Deionized Water	Deionized Water	1.0

Reddy and Chinthamreddy (1999)	To evaluate the removal of Cr, Ni and Cd in the presence of different reducing agents (humic acid, ferrous iron, or sulfide)	Kaolin and Glacial till	Cr(VI), Ni(II), and Cd(II)	Potable Water	Potable Water	1.0
Chinthamreddy and Reddy (1999)	To investigate Cr(III) oxidation and migration when the soil contained a significant quantity of Mn (1000 mg/Kg)	Kaolin and Glacial till	Cr(VI), Ni(II), and Cd(II)	Potable Water	Potable Water	1.0
Reddy et al. (1997)	To evaluate the effects of soil composition on the removal of Cr(VI)	Kaolin, Glacial till, Na-montmorillonite	Cr(VI)	Deionized Water	Deionized Water	1.0
Reddy and Parupudi (1997)	To assess the removal of multiple heavy metal contaminants from different soils	Kaolin and Glacial till	Cr(VI), Ni(II), and Cd(II)	Deionized Water	Deionized Water	1.0
Reddy and Shirani (1997)	To investigate the removal of cationic contaminants from glacial till	Glacial till	Na and Ca as surrogates	Deionized Water	Deionized Water	0.785, 1.05, 1.3, 1.57, 2.09

because chemical species have a distinct solubility, and the addition of more solution permits further contaminant solubilization into the liquid phase. Similarly, by diluting the soil with water, a more disperse or open soil matrix should result that is beneficial for contaminant migration and electroosmotic advection.

In addition, several of the previous electrokinetic investigations at UIC employed initial contaminant concentrations of 1000, 500, and 250 mg/kg for Cr, Ni, and Cd, respectively, and these concentrations were based on typical values measured at actual electroplating waste sites. It was hypothesized, however, that if lower initial concentrations were used, there would be less removal. This is because initially contaminants get sorbed to the soil surface. Only after these sorption sites have been filled, or saturated, the additional amount of contaminant remains solubilized in the liquid phase, which is conducive for easy removal.

The present study was undertaken to systematically investigate the effect of initial soil moisture content and initial contaminant concentration levels on the electrokinetic remediation of metal-contaminated soils. A total of five laboratory experiments were conducted using kaolin soil. The first three tests studied the effect of variation of initial soil moisture content under constant contaminant concentration levels and the last two tests studied the effect of variation of initial contaminant concentration levels under constant soil moisture content.

Experimental Methodology

Test Set-up

Figure 3 shows the schematic of the electrokinetic test setup (reactor) used for this study. Reddy et al. (1997) have described this setup in detail. The setup consists of an electrokinetic cell, two electrode compartments, two electrode reservoirs, a power source, and a multimeter. The Plexiglas cell had an inside diameter of 6.2 cm and a total length of 19.1 cm. Each electrode compartment consisted of a valve to control the flow into the cell, a slotted graphite electrode, and a porous stone. The electrode compartments were connected to either end of the cell using screws. The electrode reservoirs were made of 3.8 cm inner diameter Plexiglas tubes and were connected to the electrode compartments using Tygon tubing. Exit ports were created in the electrode compartments, and the tubing was attached to these ports to allow the gases generated due to the electrolysis of water to escape. The other end of these gas tubes was connected to the reservoirs to collect any liquid that was removed along with the gases. A power source was used to apply a constant voltage to the electrodes, and a multimeter was used to monitor the voltage and measure the current flow through the soil sample during the test.
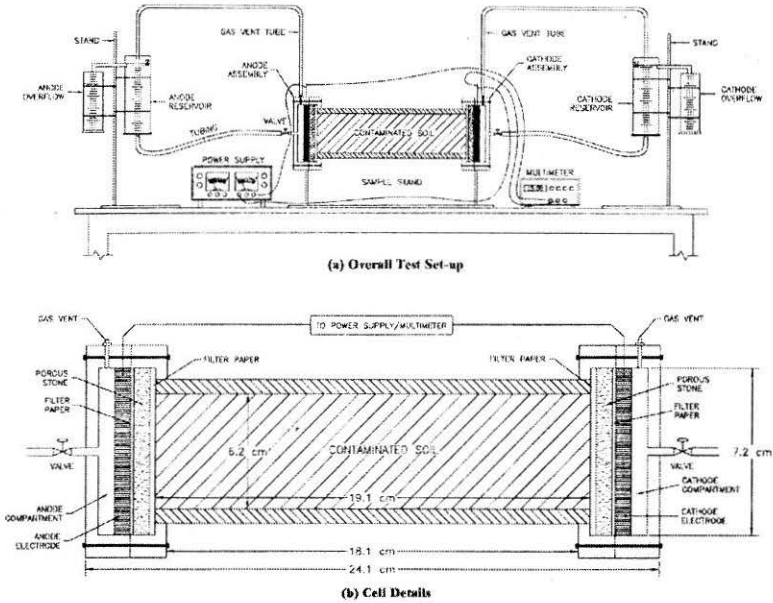
(a) Overall Test Set-up

(b) Cell Details

FIGURE 3 : Laboratory Electrokinetic Test Set-up

Test Variables

Table 2 shows the testing program and the variables used in the five different electrokinetic tests. As seen in this table, the first three tests (Tests 1 to 3) were performed using the same contaminant concentrations but at different initial moisture contents, while Tests 1, 4, and 5 were conducted using the same initial moisture content but at different levels of contaminant

TABLE 2 : Electrokinetic Testing Program

Test	Designation	Contaminant Concentration (mg/kg)	Initial Moisture Content (%)
1	EKK-1	Cr(III)-1000, Ni(II)-500, Cd(II)-250	30
2	EKK-2	Cr(III)-1000, Ni(II)-500, Cd(II)-250	50
3	EKK-3	Cr(III)-1000, Ni(II)-500, Cd(II)-250	70
4	EKK-4	Cr(III)-500, Ni(II)-250, Cd(II)-125	30
5	EKK-5	Cr(III)-250, Ni(II)-125, Cd(II)-62.5	30

TABLE 3 : Composition and Properties of Kaolin

Property	Value
Mineralogy	Kaolinite: 100%
	Muscovite: trace
	Illite: trace
Particle Size Distribution (ASTM D 422)	
Gravel	0%
Sand	4%
Silt	18%
Clay	78%
Atterberg Limits (ASTM D 2487)	
Liquid Limit	50.0%
Plastic Limit	27.4%
Plasticity Index	22.6%
Specific Gravity (ASTM D 854)	2.60
Moisture-Unit Weight Relationships	
(Harvard Miniature Compaction Test)	
Maximum Dry Unit Weight	14.4 kN/m^3
Optimum Moisture Content	27%
Hydraulic Conductivity	1.0 x 10^{-8} cm/sec
Cation Exchange Capacity (ASTM D 9081)	1.0-1.6 meq/100 g
pH (ASTM D 4972)	4.9
Organic Content (ASTM D 2974)	Near 0%
USCS Classification (ASTM D 2487)	CL

concentration. Test 4 used half the contaminant concentration of Test 1, and Test 5 used half the contaminant concentration of Test 4. All five experiments were performed using kaolin soil and deionized water as the purging solution in the anode and cathode reservoirs. The properties of the kaolin soil are shown in Table 3. In addition, for all the five experiments, the voltage gradient applied across the electrodes was 1.0 VDC/cm and the test duration was 120 hours.

Testing Procedure

Approximately 1100 g of dry soil was used for each test. Chromium chloride, nickel hydroxide, and cadmium chloride hemipentahydrate were used

as sources of Cr(III), Ni(II), and Cd(II), respectively. The amounts of chemicals to yield the desired concentrations were weighed and dissolved separately in deionized water. These contaminant solutions were then added to the soil and mixed thoroughly with a stainless steel spatula in a HDPE container. The amount of deionized water that was used was according to the testing program in Table 2. The contaminated soil was then placed in the electrokinetic cell in layers and compacted uniformly using a hand compactor. The weight of soil required in the reactor was determined and the soil-water-contaminant mixture was equilibrated for 24 hours. The electrode compartments were then connected to the electrokinetic cell. In each electrode compartment, filter papers were inserted between the electrode and the porous stone as well as between the porous stone and the soil.

The electrode compartments were connected to the anode and cathode reservoirs using Tygon tubing. The reservoirs were then filled with deionized water. Throughout the test duration, the elevation of water in both reservoirs was monitored and adjusted to prevent a hydraulic gradient forming across the specimen. The electrokinetic cell was then connected to the power supply and a voltage gradient of 1.0 VDC/cm was applied to the soil sample. The electric current and voltage across the soil sample as well as the water flow at the anode and cathode reservoirs was measured at different time periods throughout the duration of the experiment. Each test was terminated after operating for 120 hours.

At the completion of each test, aqueous solutions from the anode and cathode reservoirs and the electrode assemblies were collected and the volumes measured. Then, the reservoirs and the electrode assemblies were disconnected, and the soil specimen was extruded from the cell using a mechanical extruder. The soil specimen was sectioned into five parts at distances of 0 to 4 cm (Section 1), 4 to 8 cm (Section 2), 8 to 12 cm (Section 3), 12 to 15.5 cm (Section 4) and 15.5 to 19.1 cm (Section 5) from the anode, respectively. Each part was weighed and preserved in a glass bottle. From each soil section, 10 g of soil was taken and mixed with 10 mL of a 0.01M CaCl$_2$ solution in a glass vial. The slurry was shaken thoroughly by hand for several minutes and the solids were allowed to settle for an hour. This slurry was then used for measuring the soil pH. The pH values of the soil as well as that of the aqueous solutions from the electrodes were measured using a pH meter. The moisture content of each soil section was also determined in accordance with ASTM D2216 (ASTM, 2001).

Chemical Analyses

Representative samples from each soil section as well as from the initial soil placed in the reactor were acid digested in accordance with the USEPA chemical analysis procedure 3050 to extract the total chromium,

nickel, and cadmium (USEPA, 1989). A detailed description of the acid digestion procedure is provided in Reddy and Chinthamreddy (1999). Atomic absorption spectrophotometry (AAS) was used to measure the concentrations of chromium, nickel and cadmium in the liquid samples and the supernatant from the acid digestion procedure. These AAS analyses were done in accordance to USEPA method 7190 for chromium, 7520 for nickel, and 7310 for cadmium (USEPA, 1989).

Quality Assurance

To ensure accuracy of the test results, the following precautions were taken: (1) new electrodes, porous stones, and tubing were used for each experiment, (2) the electrokinetic cell and compartments were washed thoroughly and then rinsed first with tap water and finally with deionized water to avoid cross contamination between the experiments, (3) chemical analyses were performed in duplicate, (4) the AAS calibration was checked after testing every five samples, and (5) a mass balance was computed for each contaminant in each of the five electrokinetic tests. The final mass that was measured in the reservoirs and the treated soil, usually ranged between $\pm$ 10% of the initial mass spiked into the soil. The discrepancies in the mass balance were mainly attributed to contaminant adsorption onto the electrodes.

Results and Analysis

The results of the electrokinetic experiments were analyzed to assess the effect of variation in moisture content and contaminant concentration on contaminant migration and removal. As given in Table 2, the first three tests, Tests 1, 2, and 3, possessed different initial moisture contents, and Tests 1, 4, and 5 possessed different contaminant concentrations. The parameters analyzed included the current and cumulative electroosmotic flow values and the pH, moisture content and contaminant concentration in the soil after the experiments were terminated. The results of Tests 1, 2, and 3 were plotted together to analyze the effect of the different initial moisture contents, and the results of Tests 1, 4, and 5 were plotted together to evaluate the effect of the different contaminant concentrations.

Electrical Current

Figure 4 shows that the current values for all the tests exhibited a similar trend. Generally, the current increased rapidly during the first few hours, then decreased and converged with the other tests after about 50 hours of test operation. When water is added to the soil, the salts that are associated with the dry soil particles dissolve into the water and produce a pore solution with a high ionic strength (Mitchell, 1993). Furthermore, the dissolved ionic
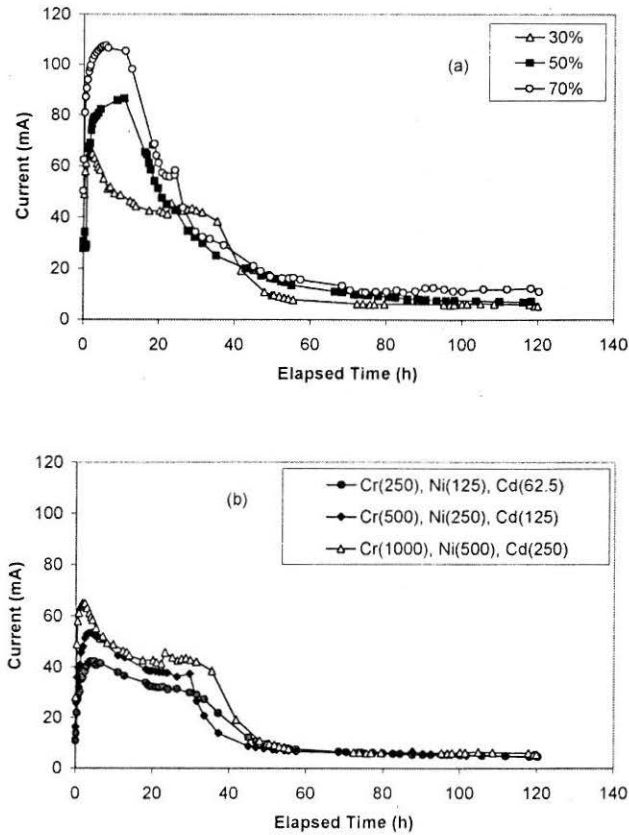
FIGURE 4 : Variation of Current with Time: (a) Tests with Different Initial Moisture Contents and (b) Tests with Different Initial Contaminant Concentrations

heavy metal contaminants that are present will also contribute to increasing the ionic strength. Initially, when the voltage gradient is first applied, the current is low because it takes time for the solution to migrate into the soil from the electrode reservoirs and for the salts and/or contaminants to dissolve. Within a few hours, however, the initial current reaches a peak value due to the strong ionic concentration. Then, the current gradually decreases because the cations and anions are electromigrating towards their respective electrode. In addition, the products of the electrolysis reactions or other chemical species may reduce the current by neutralizing the migrating ions. For instance, H^+ ions migrating towards the cathode could be neutralized by OH^- ions migrating towards the anode, thereby forming water and diluting the number of ions in solution. Time-dependent pH changes due to

electrolysis reactions could also affect the current by causing changes such as mineral dissolution, or chemical precipitation/dissolution. Unless purging solutions, which introduce additional, non-reactive, ions as charge carriers, are used, the current commonly diminishes over time (Dzenitis, 1997). Introducing electrolyte solutions can also have a negative effect on the current because it could reduce the zeta potential and the thickness of the diffuse double layer (Mitchell, 1993).

Figure 4(a) shows that the initial moisture content significantly affected the current; as the initial moisture content increased, the current increased. Increasing the moisture content might have been beneficial because the resulting soil might have been more disperse, and this should make it easier for the ionic species to migrate through the pore network. The presence of additional water molecules may also facilitate a greater amount of charged species hydration and ionic dissolution. The dilution of the ionic strength at the higher moisture content could also lead to an increased diffuse double layer thickness. In the test using the 30% moisture content, the current decreased at a slightly more gradual rate than in the experiments with higher moisture contents, and this indicates that using a lower initial moisture content may be advantageous for sustaining the current. The lower moisture content apparently hindered the electromigration of charged species, so it took additional time for the ions to reach the electrodes. Since the ions remained in the pore solution for a longer duration, the current was sustained. After around 50 hours, the current values measured were nearly the same in all the tests, and this was attributed to the electromigration and diminishing number of ionic species over time.

The current has to be proportional to the metal contaminant concentration, because the contaminant ions contribute to the ionic strength (and conductivity) of the pore solution. Figure 4(b) illustrates that, as anticipated, the current values during approximately the first 40 hours of testing corresponded to the contaminant concentrations. In other words, the test with the highest contaminant concentrations, Cr(1000), Ni(500), Cd(250), generated the highest current, the test with the middle contaminant concentrations, Cr(500), Ni(250), Cd(125), generated the mid-level current, and the test with the lowest contaminant concentrations, Cr(250), Ni(125), Cd(62.5), generated the lowest current value. However, due to the presence of other charge carrying ionic species, besides the metal contaminants, the current values were not directly proportional to the contaminant concentration. For example, as seen in Fig.4(b), the initial current values for the test with the middle contaminant concentrations, [Cr(500), Ni(250), Cd(125)], generated more than half the current of the test with the highest contaminant concentrations [Cr(1000), Ni(500), Cd(250)]. The effect of using four times the contaminant concentrations increased the initial current value only by around 50%, so the concentration effect appears to be small, and the current
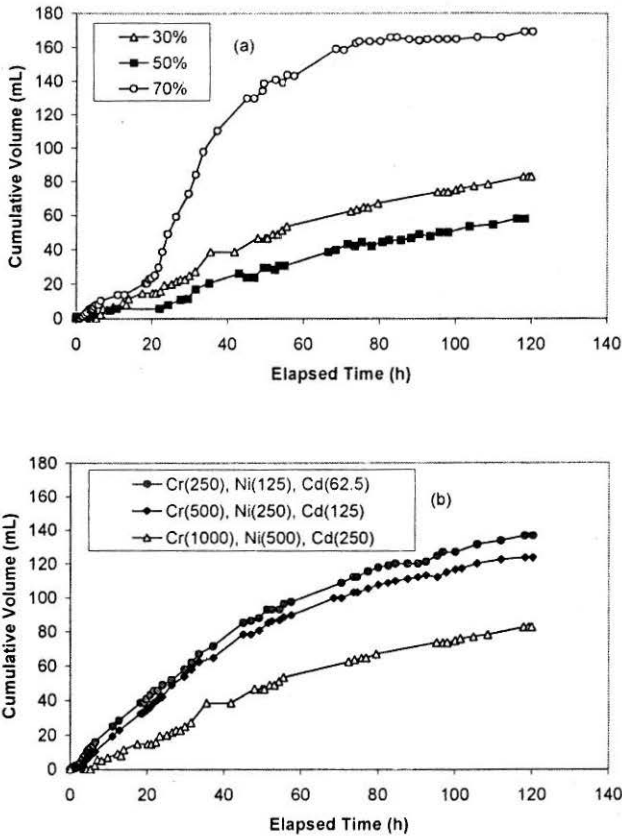
FIGURE 5 : Variation of Electroosmotic Flow with Time: (a) Tests with Different Initial Moisture Contents and (b) Tests with Different Initial Contaminant Concentrations

must have been primarily due to the salts that were originally associated with the kaolin soil particles.

Electroosmotic Flow

Figure 5 shows the electroosmotic flow measured in all five tests. The measurements showed that the current varies significantly with elapsed time, and this was attributed to the physico-chemical processes, such as the electromigration of ionic species and the electrolysis reactions. As a consequence of these changes, the surface charge of the soil particles (zeta potential) and the pore fluid properties, such as dielectric constant and viscosity, also change with time, and this influences the electroosmotic flow.

Initially, during the beginning of the test, when the current is high (electromigration is high), the transfer of momentum to the surrounding fluid molecules may be substantial. This often corresponds to a significant volume of electroosmotic flow. A high ionic strength, however, can also be detrimental for electroosmotic flow, because it reduces the thickness of the diffuse double layer and, thereby, constricts the electroosmotic flow. The charge on the soil surface must also be considered, because, as mentioned earlier, when the pH is below its ZPC, the soil particle surfaces possess a positive zeta potential and the electroosmotic flow occurs towards the anode, and when the pH is above the ZPC, the soil particles have a negative zeta potential and the electroosmotic flow occurs towards the cathode. The ZPC is also affected by the ionic strength. Although kaolin typically contains impurities that may affect its surface charge, the ZPC is usually around 4.5 (Evangelou, 1998). This ZPC value was higher than the pH for much of the soil profile (presented in the next section), but the electroosmotic flow for all the tests in this study was directed towards the cathode. This can occur because the physico-chemical properties of the soil and/or solution may vary along the profile. For example, it may be possible for a soil region possessing a large negative zeta potential and a high electrical gradient to pull the solution through a previous soil region where a small positive zeta potential and low electrical gradient exist.

Figure 5(a) shows the relationship between the elapsed time and the cumulative electroosmotic flow volume for different initial moisture contents. As seen in Fig.4(a), the current was high during the first 20 hours; but Fig.5(a) shows that the electroosmotic flow volume was actually low during this time period. It is important to note that it is the net amount of ionic migration that is significant. Thus, when an equal number of ions are migrating towards different electrodes, the transfer of momentum is nearly the same in each direction, so the net result is a low electroosmotic flow. In addition, the high conductivity near the start of the experiments may have reduced the thickness of the diffuse double layer and constricted the electroosmotic flow. After the first 20 hours, however, it appears as if the physico-chemical changes, such as the reduction in conductivity due to electromigration and/or the increasing concentration and migration of the products from the electrolysis reactions, caused the flow to increase. As explained earlier, the H^+ ions that are generated by the electrolysis reaction at the anode are transported into the soil by electromigration and electroosmosis, and this lowers the pH in the soil, especially near the anode. As the pH in the soil drops in this region, metal species may dissolve because many metals are more soluble under low pH conditions. Consequently, the cationic metal species will electromigrate towards the cathode, but near the cathode, they may adsorb and/or precipitate. This is because many metals form precipitates under high pH conditions, for instance, Cr(III), Ni(II), and Cd(II) ions may form

precipitates, such as $Cr(OH)_3$, $Ni(OH)_2$, and $Cd(OH)_2$, respectively, under high pH conditions.

The three experiments that employed different initial moisture contents generated a low amount of flow during the first 20 hours. Hence, apparently, the initial moisture content was not a significant factor during this stage. As time passed, however, the test employing the 70% moisture content clearly generated a greater volume of flow compared to the other two tests. Figures 5(a) and 4(a) show that the flow rate in the test using the 70% moisture content was the highest when the current was decreasing, approximately between the elapsed time of 20 to 40 hours. It is possible that this reduction in current corresponded to an expansion of the diffuse double layer (or an increase in the magnitude of the zeta potential), and this facilitated a greater electroosmotic flow. In addition, due to the precipitation of metal species near the cathode, a steep electrical gradient may form adjacent to the cathode, and the combination of the steep electrical gradient and thick diffuse double layer are beneficial for electroosmotic flow. Since the metals may precipitate near the cathode, the amount of electromigration in the cathode region may be limited. But, due to the steep electrical gradient and thick diffuse double layer, a small amount of electromigration in this region could generate a significant volume of electroosmotic flow (Dzenitis, 1996). It seems that when the high, 70%, moisture content was used, the flow was greater because electromigration and fluid transport were facilitated by the more disperse clay particles.

As seen in Table 3, the properties of the kaolin soil indicate that the plastic limit is about 27 and the liquid limit is 50. These values represent respectively the moisture contents at which the soil changes from a semi-solid to a plastic state and from a plastic state to a liquid state that can produce a viscous flow (Holtz and Kovacs, 1981). Therefore, at the two lower moisture contents, 30% and 50%, the soil condition was similar because it was in a plastic state, but at the higher, 70%, moisture content, the soil behavior changed and became more like a thick liquid or sludge. Figure 5(a) shows that the tests using the lower moisture contents, 30% and 50%, generated moderate amounts of electroosmotic flow, while the test using the 70% moisture content generated a substantially higher flow. These results suggest that soils that have moisture contents that exceed the liquid limit may generate a higher flow, whereas soils in the plastic state may generate lower amounts of flow.

From Figures 4(b) and 5(b), it is evident that a higher current production corresponded to a lower electroosmotic flow. It seems that as the contaminant concentration increased, the thickness of the diffuse double layer reduced, and this constricted the electroosmotic flow. It is also possible to some extent that contaminant precipitation near the cathode contributed to clogging the pore spaces diminishing the number of flow paths. It is

interesting to note that the two tests with low contaminant concentrations generated comparable amounts of flow, while the test with the highest contaminant concentration produced a significantly lower amount of flow. These results suggest that the ionic concentration effect was not linear and there may be a threshold value where the effects on the diffuse double layer become significant and the flow is considerably reduced.

Soil pH

As discussed earlier, the electrolysis of water results in the formation of H^+ ions (low pH solution) at the anode and OH^- ions (high pH solution) at the cathode, and, primarily due to electromigration, these ions tend to migrate towards the oppositely charged electrode(s). For a low acid buffering clay soil, like kaolin, the acidic solution generated at the anode typically migrates through the soil towards the cathode, thereby lowering the pH along most of the soil profile. Compared to H^+ ions, OH^- ions have larger ionic radii and a lower mobility and hence the H^+ ions usually migrate faster through the soil (Acar and Alshawabkeh, 1993). The difference in the rate of the electrolysis reactions and the contribution of electroosmosis may also benefit H^+ transport.

Figure 6 shows the normalized distance from the anode versus the soil pH for all the tests. The normalized distance is defined as the distance to the specific location from the anode divided by the total distance from the anode to the cathode. The initial pH of the soil was 3.55, 3.64 and 3.57 for the tests using the 30, 50, and 70% moisture content, respectively. Figure 6(a) illustrates that an acidic front of solution, which was generated by the electrolysis reaction at the anode, migrated from the anode towards the cathode, and this solution significantly lowered the pH through the first three soil sections from the anode. Conversely, it is also evident from this figure that an alkaline solution, generated by the electrolysis reaction at the cathode, migrated towards the anode and increased the pH in the soil region nearest to the cathode. By analyzing Figures 5(a) and 6(a), it can be seen that the test with 70% moisture content produced the highest cumulative flow, and this test had the lowest pH in the soil adjacent to the cathode. If there is a high electroosmotic flow towards the cathode, the pH should be lower near the cathode, because the direction of electroosmotic flow was from the anode towards the cathode, thereby opposing the electromigration of OH^- ions towards the anode. Moreover, when the concentration of H^+ ions increases near the cathode, more OH^- ion neutralization may occur. The tests with the lower moisture contents, 30 and 50%, produced a similar amount of electroosmotic flow, and they possessed similar pH profiles. The lower flow in the test with 50% moisture content might have allowed greater OH^- penetration, because the second soil section from the cathode possessed a slightly higher pH than in the other two tests. The greatest variation between
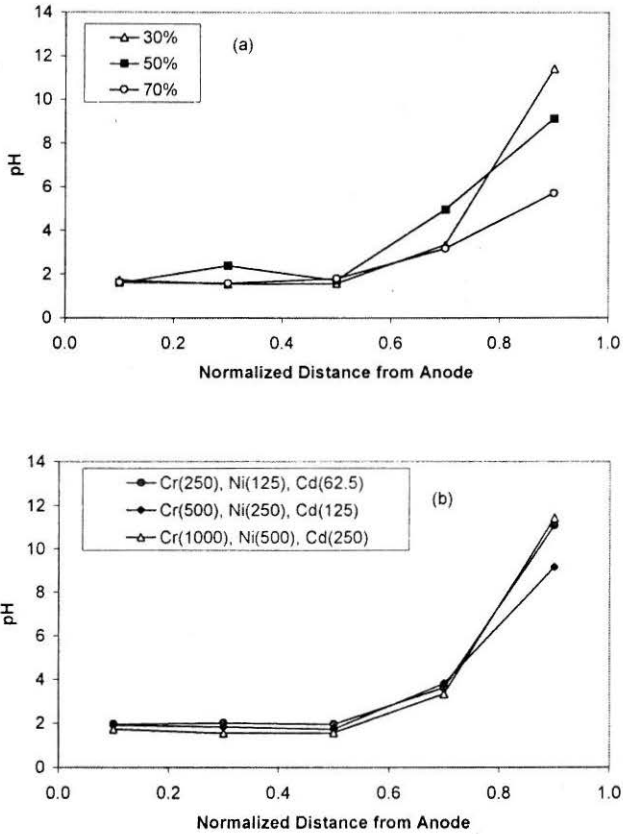
FIGURE 6 : Variation of Soil pH with Distance: (a) Tests with Different Initial Moisture Contents and (b) Tests with Different Initial Contaminant Concentrations

the three tests occurred in the soil section adjacent to the cathode, and this mainly appeared to be a consequence of the variable electroosmotic flow rates of these experiments. These results indicate that the initial moisture content does have an effect on the pH, but the effect was minor and was mostly confined to the region nearest to the cathode.

As seen in Fig.6(b), the pH profiles for the three tests using different concentration levels were almost identical. As seen in Fig.5(b), the two tests using the lower concentrations had nearly the same electroosmotic flow and hence similar pH profiles would be expected. However, it was somewhat surprising that the test with the highest concentration and the lowest amount of flow did not possess soil sections near the cathode with a higher alkalinity.
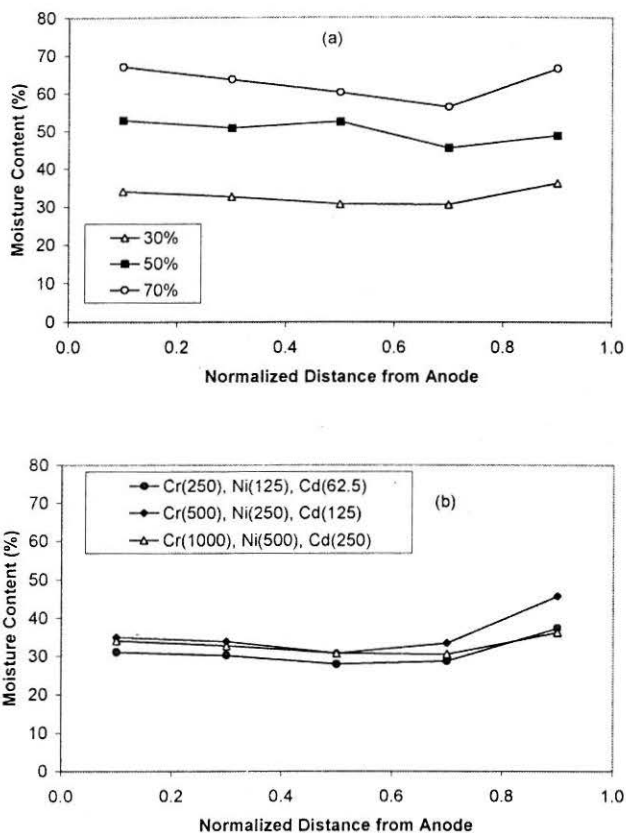
FIGURE 7 : Variation of Moisture Content with Distance: (a) Tests with Different Initial Moisture Contents and (b) Tests with Different Initial Contaminant Concentrations

Apparently, since all these tests were conducted at the low, 30%, moisture content, the soil might have been denser. So it was difficult for the larger and less mobile OH^- ions to penetrate into the soil from the cathode. Clearly though, for all three tests, the pH rose considerably in the soil section adjacent to the cathode. The small amount of variation in these experiments indicates that compared to the initial moisture content, the initial contaminant concentrations in the soil do not significantly affect the pH.

Moisture Content

As shown in Fig.7(a), the moisture content of the soil at the conclusion of testing was comparable to the initial moisture contents of 30, 50, and

70%. Hence it appears that the electrokinetic process does not significantly alter the moisture content. Slight changes in moisture contents are evident which can be attributed to the variations in the electroosmotic flow that occurred as a result of the changes in parameters such as the ionic strength, conductivity, and/or electrical gradient. These results suggest that the electroosmotic flow might not be uniform and there might be changes in pore pressures (Eykholt, 1997). Nevertheless, it appears that the soil moisture content remained fairly consistent and comparable to the initial moisture content. As implied earlier, it is possible that regions where the electroosmotic flow was high created a pressure gradient so that the solution was pulled from regions where the electroosmotic flow was lower. Since the solution was continuously transported through the soil, the moisture content did not substantially deviate from the initial moisture content. These results suggest that the initial moisture content stays approximately constant throughout testing, and this is important because, as seen previously with the current and electroosmotic flow, the initial moisture content can significantly affect the electrokinetic remediation processes.

Figure 7(b) shows the soil moisture content profile at the end of testing for the experiments employing different contaminant concentrations. As seen in this figure, for all three tests, the moisture content remained fairly close to the initial moisture content of 30%. It is evident, however, that the moisture content increased to a minor extent in some regions, especially in the soil sections adjacent to the cathode, and this is primarily attributed to the variations in the electroosmotic flow that occurred. The close resemblance of the moisture content results from the different experiments indicates that the variation in contaminant concentration had a minimal influence on the soil moisture content during the electrokinetic process.

Contaminant Migration and Removal

Based on their solubility when they are alone in water (i.e., without soil) Cr(III) has a low solubility and starts precipitating as $Cr(OH)_3$ at pH value of approximately 5 (Fendorf, 1995), Ni(II) starts precipitating as $Ni(OH)_2$ at pH values of 6 to 7, and Cd(II) starts precipitating as $Cd(OH)_2$ at pH value of 7 to 8, (Chinthamreddy, 1999). Therefore, the Cr(III), Ni(II) and Cd(II) ions that were electromigrating towards the higher pH region close to the cathode, might precipitate in the order of $Cr(OH)_3$, $Ni(OH)_2$, and then $Cd(OH)_2$. This analysis, however, has been greatly simplified since many additional factors such as the synergistic effects of the co-contaminants, the effects of the soil and contaminant adsorption, and the electrokinetic effects have not been considered.

Figure 8 illustrates the contaminant concentrations that were measured along the soil profile after the electrokinetic process was complete for the
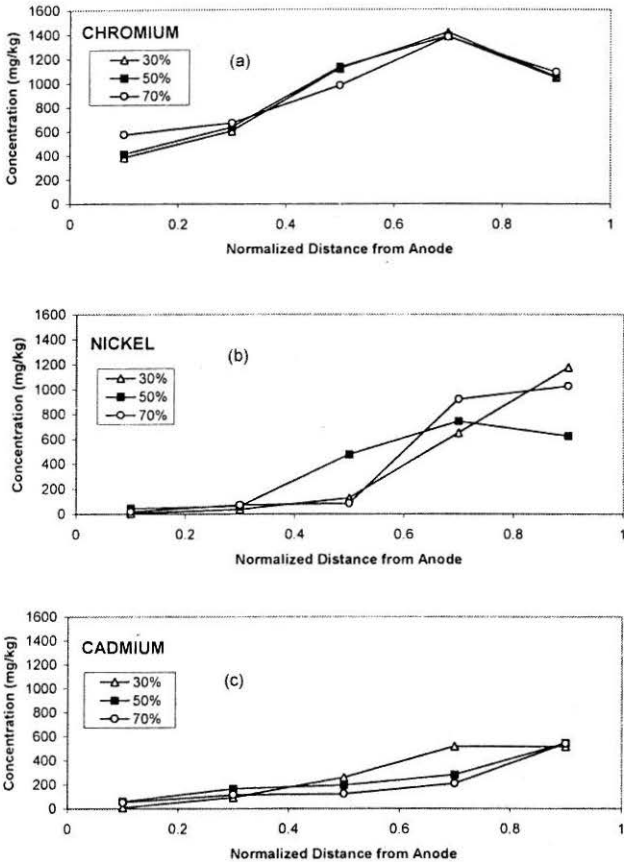
FIGURE 8 : Variation of Contaminant Concentrations with Distance for Different Initial Moisture Contents

tests with different moisture contents. It should be noted that the initial contaminant concentrations for the all these tests were Cr(1000), Ni(500), and Cd(250). In the concentration profile for chromium, Fig.8(a), it can be observed that the Cr(III) concentration decreased in the first two soil sections from the anode and it increased considerably in the second soil section from the cathode. As seen in Fig.6(a), the pH values in the soil regions near the anode were below 3. So some of the chromium in this region was probably soluble and was present in the Cr(III) cationic form. In this form, the cations could migrate by electromigration, and possibly by electroosmosis to some extent, towards the cathode. Figure 6(a) shows that the pH increased to around 4 to 5 in the second soil section from the cathode and so it is likely that the Cr(III) species precipitated in that region as $Cr(OH)_3$. Since the

concentration profiles are similar for all three water contents, it appears that the chromium migration was not significantly affected by the initial moisture content.

Figures 8(b) and 8(c) indicate that the Ni(II) and Cd(II) cations behaved in a manner that was similar to Cr(III) in that they migrated towards the cathode and then precipitated in the soil regions near the cathode. These results suggest that the Ni(II) and Cd(II) migration was more extensive than Cr(III) because nearly double the initial concentrations were present in the soil section adjacent to the cathode, and very low concentrations were present in the soil sections near the anode. This was especially obvious for the Ni(II) contaminant since the first two soil sections from the anode had negligible Ni concentrations for all three moisture content tests. Although some variation in the Ni(II) and Cd(II) migration occurred between the tests using different moisture contents, a noticeable trend was not evident. Apparently, the initial moisture content did not significantly affect the contaminant migration or removal. In addition, by observing Fig.5(a) and Fig.8, it can be seen that the higher electroosmotic flow that occurred in the test with a moisture content of 70% did not seem to benefit contaminant migration. So the influence of electroosmotic flow was concluded to be minimal.

Figure 9 shows the normalized concentrations of the Cr(III), Ni(II), and Cd(II) contaminants that were present in the soil for the tests conducted with different initial contaminant concentration levels. The normalized concentration is defined as the final measured concentration divided by the initial concentration. Figures 8 and 9 indicate that the cationic contaminants migrated towards the cathode. As illustrated in Fig.9(a), compared to the initial concentration in the soil, the Cr(III) was removed similarly for all three experiments, even though it was initially present at different concentrations. This suggests that at the levels tested, the initial concentration had minimal effect on Cr(III) migration. For instance, for all the three experiments, about half the initial Cr(III) was measured in the soil section adjacent to the anode, while about 1.5 times the initial amount was measured in the second soil section from the cathode.

Figures 9(b) and 9(c) indicate that the initial concentration had an effect on the removal of the Ni(II) and Cd(II) contaminants, respectively. In both the cases, the test with the highest contamination levels, Cr(1000), Ni(500), Cd(250), exhibited less contaminant migration along the soil profile. In Fig.9(b), this was observed only at the second section from the cathode for Ni(II). But, in Fig.9(c), it is higher for all the three middle soil sections for Cd(II). Although these differences could be due to the different initial Ni(II) and Cd(II) contaminant concentrations, different adsorption affinities of heavy metals for soils might have also contributed to these differences (Yong, 1995). It appears that in the test with the highest contaminant concentrations,
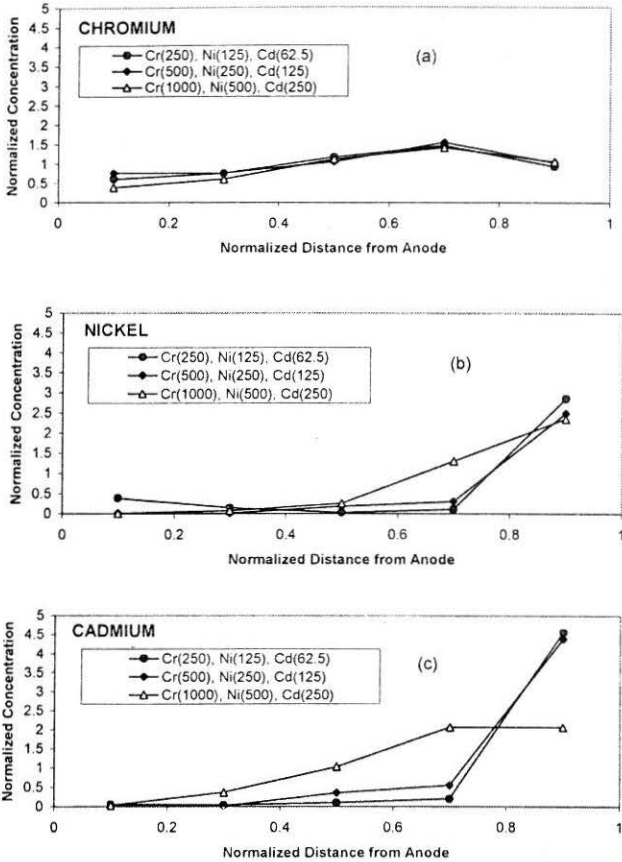
FIGURE 9 : Variation of Normalized Contaminant Concentrations with Distance for Different Initial Contaminant Concentrations

the Cd species may have had a stronger adsorption to the kaolin than the Ni species. The comparatively low removal of Cr(III) indicates that this contaminant could also have a strong affinity for adsorbing to kaolin, but $Cr(OH)_3$ starts precipitating at relatively low pH values of 3 to 4. The two tests with lower initial contaminant concentrations showed a high amount of Ni(II) and Cd(II) contaminant migration except for the soil section adjacent to the cathode, where they most likely precipitated. This is evident because, as seen in Fig.6(b), the pH increased significantly in that region. In Fig.5(b), it was seen that the two tests with the lower contaminant concentrations produced a higher electroosmotic flow. So this might have also contributed to the higher Ni(II) and Cd(II) contaminant migration in these tests. Clearly though, Figs.9(b) and 9(c) indicate that contaminant migration and removal

can be affected by the initial contaminant concentrations, and contaminant migration was lower when the metals were present in higher concentrations.

Summary and Conclusions

This paper provides an overview of electrokinetic remediation and presents the results of five bench-scale electrokinetic experiments performed to investigate the effects of initial moisture contents and initial contaminant concentrations on contaminant migration and removal in soils. The first three experiments were conducted using different moisture contents of 30, 50 and 70%, with same heavy metal contaminants, Cr(III), Ni(II), and Cd(II), at contaminant concentrations of 1000, 500, and 250 mg/kg, respectively. Then two additional experiments were conducted at lower Cr(III), Ni(II) and Cd(II) concentrations of 500, 250, and 125 mg/kg and 250, 125, and 62.5 mg/kg, respectively. But these tests used the same moisture content of 30%.

The results revealed that the electrical current and electroosmotic flow increased considerably when the soil contained a higher, 70%, moisture content, but the moisture content effect on heavy metal contaminant migration and removal appeared to be minimal. Furthermore, these experiments indicated that the moisture content remains nearly the same during the electrokinetic process although slight changes were evident. These slight changes were attributed to minor variations in the electroosmotic flow, which were, in turn, the result of the physico-chemical reactions and electrokinetic transport mechanisms that were occurring.

The results showed that soils possessing higher contaminant concentrations could have a lower percentage of contaminant migration and removal. Although similar amounts of removal occurred for the Cr(III) contaminant, Ni(II) and Cd(II) migration was evidently inhibited when the three co-contaminants were present in the test using the highest, 1000, 500, and 250 mg/kg, concentrations, respectively. These results indicated that there might have been a greater adsorption affinity for Cd(II) to kaolin than for Ni(II) to kaolin, but this was observed only when the three contaminants were present at the highest concentrations. Clearly, the effect of the initial contaminant concentration depends on the type and concentration of the contaminants that are present.

Overall, it was concluded that the initial moisture content affects the electrokinetic process but it does not significantly influence the migration and removal of heavy metal contaminants, and the results indicate that the initial contaminant concentrations affect migration and removal, but the effect is dependent on the type and concentration of the heavy metal contaminants that are present.

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