

# Complicating Factors of Using Ethylenediamine Tetraacetic Acid to Enhance Electrokinetic Remediation of Multiple Heavy Metals in Clayey Soils

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**Abstract:** Batch and electrokinetic experiments were conducted to investigate the removal of three different heavy metals, chromium(VI), nickel(II), and cadmium(II), from a clayey soil by using ethylenediamine tetraacetic acid (EDTA) as a complexing agent. The batch experiments revealed that high removal of these heavy metals (62–100%) was possible by using either a 0.1 M or 0.2 M EDTA concentration over a wide range of pH conditions (2–10). However, the results of the electrokinetic experiments using EDTA at the cathode showed low heavy metal removal efficiency. Using EDTA at the cathode along with the pH control at the anode with NaOH increased the pH throughout the soil and achieved high (95%) Cr(VI) removal, but the removal of Ni(II) and Cd(II) was limited due to the precipitation of these metals near the cathode. Apparently, the low mobility of EDTA and its migration direction, which opposed electroosmotic flow, prevented EDTA complexation from occurring. Overall, this study found that many complicating factors affect EDTA-enhanced electrokinetic remediation, and further research is necessary to optimize this process to achieve high contaminant removal efficiency.

**DOI:** 10.1061/(ASCE)0733-9372(2004)130:11(1357)

**CE Database subject headings:** Abatement and removal; Cadmium; Chromium; Chelating agents; Electrokinetics; Heavy metals; Soil treatment; Soil pollution.

## Introduction

Mostly due to mining operations and the improper disposal practices of various metal application industries (e.g. electroplating, textile, leather tanning), numerous sites have been contaminated with heavy metals, such as lead, chromium, nickel, cadmium, mercury, and arsenic (USEPA 1995). Heavy metals pose substantial risks to public health and the environment, so the prompt remediation of these contaminated sites is clearly warranted. A variety of treatment techniques, such as soil washing/flushing and solidification and stabilization, have been used for heavy metal-contaminated soils, but when the subsurface possesses considerable quantities of clayey soils, these conventional methods usually become costly and/or success is limited. However, electrokinetic remediation has great promise for these situations.

Electrokinetic remediation, or simply electrokinetics, first involves the installation of wells/drains that surround the contami-

nated region. Electrodes are then inserted into the wells/drains, and/or into the soil, and a low electric potential is applied across the electrodes. The electric potential induces contaminant transport mechanisms that cause the heavy metals to migrate toward the electrodes where they can be extracted from the subsurface. Compared to most conventional remediation techniques, electrokinetics is safe, economical, and easy to implement, but the complicated physico-chemical processes that occur largely govern remedial efficiency. Therefore, to accomplish high contaminant removal, it is crucial to have a fundamental understanding of the physico-chemical processes involved, and several previous studies, such as those by Gray and Mitchell (1967), Eykholt (1992), Acar and Alshawabkeh (1993), Eykholt and Daniel (1994), and Acar et al. (1995) have focused on the different factors and processes that affect electrokinetic remediation.

During electrokinetics, the electrolysis of water occurs at the electrodes, generating oxygen gas and hydrogen ( $H^+$ ) ions (low pH solution) at the anode and hydrogen gas and hydroxyl ( $OH^-$ ) ions (high pH solution) at the cathode



Furthermore, the electric potential induces several contaminant transport mechanisms, such as electromigration, electroosmosis, electrophoresis, and diffusion. Electromigration refers to the transport of ionic species present in the pore fluid, and this is the main mechanism by which the electrical current flows through the soil. Electromigration also includes the movement of  $H^+$  and  $OH^-$  ions produced by electrolysis toward the oppositely charged electrode. Electroosmosis describes the bulk movement of the pore

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Note. Associate Editor: Mark J. Rood. Discussion open until April 1, 2005. Separate discussions must be submitted for individual papers. To extend the closing date by one month, a written request must be filed with the ASCE Managing Editor. The manuscript for this paper was submitted for review and possible publication on August 6, 2002; approved on October 3, 2003. This paper is part of the *Journal of Environmental Engineering*, Vol. 130, No. 11, November 1, 2004. ©ASCE, ISSN 0733-9372/2004/11-1357-1366/\$18.00.

fluid. Electroosmotic flow is produced because locally existing excess ions migrate in a plane parallel to the soil particle surface toward the oppositely charged electrode, and, as they migrate, they transfer momentum to the surrounding fluid molecules via viscous forces (Eykholt 1992). Finally, electrophoresis describes the transport of colloidal size particles through the pore fluid, and diffusion refers to the transport of species due to concentration gradients.

It should be noted that the contaminant transport occurs primarily through the pore fluid, so the electrokinetic remediation process is not effective unless the contaminants are soluble, or they are converted to a soluble form. Thus, physico-chemical processes, such as acid/base reactions, adsorption/desorption, precipitation/dissolution, and oxidation/reduction greatly affect electrokinetic remediation. Furthermore, these processes are highly dependent on the type and speciation of the particular metals that are present. For example, chromium species essentially exist in two forms, hexavalent chromium (Cr(VI)) and trivalent chromium (Cr(III)). Cr(VI) usually exists as anionic complexes, while Cr(III) exists as cationic complexes and/or cationic hydroxo complexes. Thus, during electrokinetic remediation, Cr(VI) migrates toward the positively charged electrode (anode) whereas Cr(III) migrates toward the negatively charged electrode (cathode) (Reddy et al. 2001a; Reddy and Chinthamreddy 2003a). The adsorption/desorption chemistry of heavy metals may also be quite different. For instance, the adsorption of Cr(VI) in soil is negligible at high pH; however, the adsorption/precipitation of Cr(III) in soil is significant at high pH. Other heavy metals, such as nickel (Ni(II)) and cadmium (Cd(II)) commonly exist as cations and cationic complexes, and they migrate toward the cathode during electrokinetic treatment. When heavy metals such as Cr(VI), Ni(II) and Cd(II) coexist, there may also be synergistic effects on their electrokinetic remediation (Reddy et al. 2001b).

In low acid buffering soils such as kaolin, a low pH (2.0–3.0) solution develops due to the electrolysis reaction at the anode, and this solution is transported into the soil in the direction of the cathode by electromigration. When the electroosmotic flow is in the same direction, it assists in transporting the low pH solution toward the cathode. Since many cationic heavy metals, such as Ni(II) and Cd(II) species are soluble under low pH conditions, these metals are dissolved as the pH reduces, and they migrate with the H<sup>+</sup> ions toward the cathode. However, the high pH (8.0–12.0) conditions that develop as a result of the electrolysis reaction at the cathode can cause the metals to adsorb to the soil or precipitate as hydroxides, so the actual contaminant removal is usually insignificant (Reddy et al. 1997; Reddy and Parupudi 1997; Reddy and Chinthamreddy 2003a). Consequently, various researchers have tried adding different solutions, such as weak acids and/or complexing or chelating agents, to enhance metal solubility near the cathode and improve remedial efficiency (Eykholt 1992; Reed et al. 1995; Yeung et al. 1996; Wong et al. 1991; Reddy and Chinthamreddy 2003b). Among these solutions, ethylenediamine tetraacetic acid (EDTA) has shown great promise. Ethylenediamine tetraacetic acid has been also employed to treat heavy metal-contaminated soil using conventional soil flushing/washing, as in studies by Heil et al. (1999), Lo and Yang (1999), Tambouris et al. (1999), Kontopoulos et al. (2000), and Sun et al. (2001).

Ethylenediamine tetraacetic acid, as shown in the following diagram, is hexadentate ligand that has six electron pairs (four carboxyl (COO<sup>-</sup>) and two nitrogen (N) donor groups) that are capable of being shared with one or more metal cations (Lindsay 1979). With increasing pH, there is a greater tendency for the H<sup>+</sup>

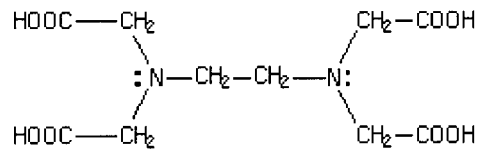
**Table 1.** Stability Constants of Metal–Ethylenediamine Tetraacetic Acid (EDTA) Complexes (Stumm and Morgan 1996)

Ion	Complex	Stability constant
Cd <sup>2+</sup>	CdL	18.2
	CdHL	21.5
Ni <sup>2+</sup>	NiL	20.4
	NiHL	24.0
	NiOHL	21.8
H <sup>+</sup>	HL	11.12
	H <sub>2</sub> L	17.8
	H <sub>3</sub> L	21.04
	H <sub>4</sub> L	23.76
	H <sub>5</sub> L	24.76

<sup>a</sup>Note: L=EDTA ligand.

ions to dissociate from the carboxyl groups, and when all the H<sup>+</sup> ions dissociate, EDTA exists as an anionic ligand (L<sup>4-</sup>). As EDTA is added to a soil, numerous cationic species may compete to reach thermodynamic equilibrium and to develop the most stable metal chelate(s). Lindsay (1979) reported that the reactions of metals with EDTA cause solid phases to dissolve and exchangeable ions to dissociate from the soil to replenish the ions that are chelated. Stability constants are helpful and provide an indication of the speciation of the chelated species that are present (Table 1), but, as suggested earlier, during electrokinetics, nonequilibrium conditions exist due to the multiple physico-chemical processes that are occurring.

**Ethylenediamine tetraacetic acid (EDTA) – H<sub>4</sub>L<sup>0</sup>**



To date, there have been very few investigations concerning the EDTA-enhanced electrokinetic remediation of heavy metal-contaminated soils, and the studies that have been performed have been limited in scope (Reed et al. 1995; Yeung et al. 1996; Wong et al. 1997). Most of the previous studies have dealt with only a single contaminant, such as lead, whereas contaminated sites generally have multiple contaminants. Furthermore, some of these previous studies have used sandy soils, but, due to their low hydraulic conductivity, soils with a large fraction of silt or clay are generally much more difficult to remediate.

This present laboratory research study investigates the EDTA-enhanced electrokinetic remediation of multiple heavy metal contaminants, Cr(VI), Ni(II), and Cd(II), from a clayey soil, kaolin. Initially, batch experiments (soil washing) were performed to assess the capability of EDTA to solubilize the heavy metals at different concentrations and under different pH conditions. Then, five bench-scale electrokinetic experiments were conducted to assess the use of EDTA as a purging solution. The batch and electrokinetic experimental results helped to assess the complicated physico-chemical processes affecting EDTA transport into soil under an electric potential and its extent of complexation with metal contaminants.

**Table 2.** Properties of Kaolin Soil

Property	Value
Mineralogy	Kaolinite:100% Muscovite: trace Illite: trace
Particle size distribution, (%) (ASTM D 422)	
Gravel	0
Sand	0
Silt	10
Clay	90
Hydraulic conductivity (cm/s) (ASTM D 2434)	$1.0 \times 10^{-8}$
Cation exchange Capacity, meq/100 g (ASTM D9081)	1.0–1.6
pH (ASTM D 4972)	4.5–5.0
USCS classification (ASTM D 2487)	CL

Note: ASTM=American Society for Testing and Materials; USCS =Unified Soil Classification System; and CL=Clay.

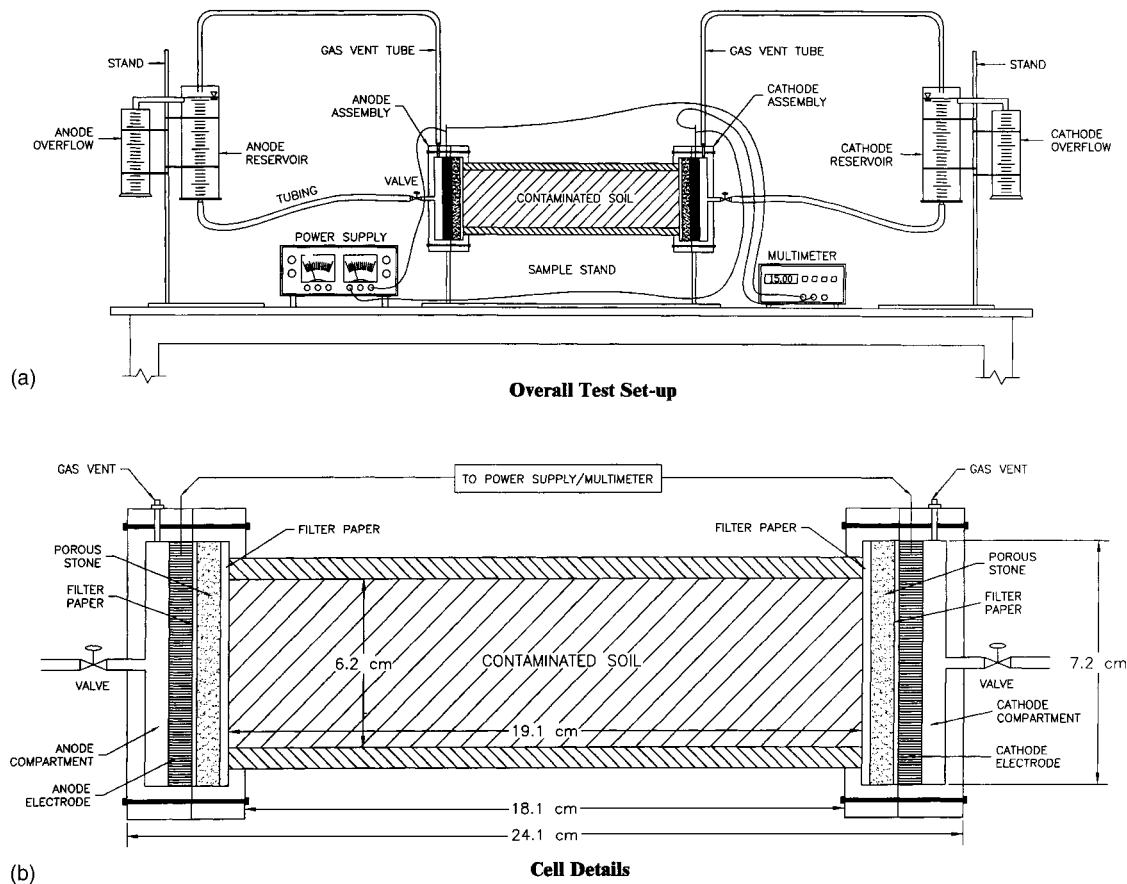
## Materials and Methods

### Materials

Table 2 summarizes the composition and properties of the kaolin used in this investigation. Kaolin is commonly used in electrokinetic experiments because, compared to most clayey soils, it has a uniform chemical composition and its soil properties have been characterized in detail (Eykholt 1992).

The chemical compounds used to artificially contaminate (spike) kaolin with heavy metals included potassium dichromate ( $K_2CrO_4$ , certified ACS) for Cr(VI), nickel chloride ( $NiCl_2 \cdot 6H_2O$ , technical Fisher Chemical) for Ni(II), and cadmium chloride ( $CdCl_2 \cdot 2.5H_2O$ , Certified ACS) for Cd(II). The solutions that were used included deionized water (pH=5.7, redox=199 mV, electrical conductivity=0.1  $\mu S/cm$ ), and 0.1 M and 0.2 M EDTA, disodium salt dihydrate ( $(HO_2CCH_2)_2NCH_2CH_2N(CH_2CO_2Na)_2 \cdot 2H_2O$ , Certified ACS). For the batch experiments, the pH of the EDTA solutions was adjusted using either dilute nitric acid ( $HNO_3$ ) or sodium hydroxide (NaOH).

A schematic of the electrokinetic test setup used for this study is shown in Fig. 1. The setup basically consisted of an electrokinetic cell, two electrode compartments, two electrode reservoirs, a power supply, and a multimeter. The cell had an inside diameter of 6.2 cm and a length of 19.1 cm, and the cell and the electrode reservoirs were made of Plexiglas. Each electrode compartment consisted of a control valve, a slotted graphite electrode, a porous stone, and a filter paper. Gas vents were used to allow the gases



**Fig. 1.** Bench-scale electrokinetic test setup

**Table 3.** Experimental Program

Test	Contaminant concentration (mg/kg)			Voltage gradient (VDC/cm)	Anode solution	Cathode solution	Duration (days)
	Cr(VI)	Ni(II)	Cd(II)				
1	1,000	500	250	1	De-ionized water	De-ionized water	4
2	1,000	500	250	1	Deionized water	0.2 M EDTA	4
3	1,000	500	250	1	0.1 M NaOH	0.2 M EDTA	4
4	1,000	500	250	1	0.5 M NaOH	0.2 M EDTA	11
5	1,000	500	250	2	0.5 M NaOH	0.2 M EDTA	2

Note: VDC=volts of direct current; and EDTA=ethylenediamine tetraacetic acid.

produced from the electrolysis reactions to escape. The electrode reservoirs were graduated to facilitate the measurement of flow into or out of the cell.

### Testing Procedure

For all the tests, the kaolin was spiked with Cr(VI), Ni(II), and Cd(II) at concentrations of 1,000, 500, and 250 mg/kg, respectively. These concentrations were selected based on the typical concentrations found at sites contaminated by electroplating wastes. The required amounts of the chemical compounds that would yield the desired concentrations of Cr(VI), Ni(II), and Cd(II) were weighed and dissolved individually in deionized water. Approximately 1.4 kg of dry kaolin soil was used for each electrokinetic experiment. The contaminant solutions were added and mixed with the dry kaolin in a high-density polyethylene container using a stainless steel spatula. A total volume of 490 mL of de-ionized water was used to simulate typical soil moisture condition of 35%. After spiking, the soil was placed in glass bottles and stored in the refrigerator for at least 1 week to equilibrate. The pH, redox potential, and electrical conductivity of the soil was measured both before and after equilibration. A sample from each batch of spiked soil was retained for chemical analysis and the determination of the initial heavy metal concentrations.

For the batch experiments, 5 g of the spiked kaolin was added to 10 mL of the EDTA solution, and the flasks were then placed on a shaker table for 24 h. Duplicate flask experiments were conducted to verify the reproducibility of the testing procedure. After shaking, each mixture was centrifuged at 4,000 rpm for 30 min, and the supernatant was collected for chemical analysis.

For the electrokinetic tests, the spiked kaolin was tamped into the cell using a hand compactor so that the amount of void space was minimized. The electrode compartments were then connected to the anode and cathode reservoirs by chemically resistant Tygon tubing. The electrode compartments and reservoirs were then filled with the desired electrode solution. The elevation of the electrode solution in both reservoirs was kept at nearly the same level to minimize the hydraulic gradient. The electrokinetic cell was then connected to the power supply and the required constant voltage gradient was applied. The electric current across the soil sample as well as the electroosmotic flow, pH, redox potential and electrical conductivity in both the anode and cathode reservoirs was measured periodically throughout the duration of the experiment. The anode and the cathode reservoirs were emptied and replenished occasionally to ensure that the contaminant that migrated into these reservoirs did not diffuse back into the soil. Solution samples were retained for chemical analysis.

At the end of each electrokinetic test, the solutions were drained and collected from the anode and cathode reservoirs as

well as the electrode compartments. The volume of these solutions was then measured. The electrokinetic reactor was disassembled, and the soil specimen was extruded from the cell using a mechanical extruder. The soil specimen was sectioned into five parts, and each part of the soil was weighed and subsequently preserved in a glass bottle for chemical analysis.

### Testing Program

A total of five electrokinetic experiments were conducted and the experimental variables are shown in Table 3. As seen in this table, the first experiment used deionized water at both the anode and cathode, and this experiment served as a baseline test for comparison purposes. The second test was identical to the first, but this test used a 0.2 M EDTA solution at the cathode to enhance heavy metal dissolution and removal. The third and fourth tests were identical to the second test, but these tests employed a 0.1 and 0.5 M NaOH solution, respectively, at the anode to increase the pH throughout the specimen. As shown in Table 1, the stability constant for H<sup>+</sup>-EDTA complexes can be comparatively high, so under low pH conditions, it may be thermodynamically favorable for EDTA anions to dissociate with Ni(II) or Cd(II) and complex with H<sup>+</sup>. Thus, it was hypothesized that increasing the pH at the anode would benefit the electromigration of Ni-EDTA and/or Cd-EDTA anionic complexes toward the anode. Furthermore, Cr(VI) species may adsorb to the soil under low pH conditions, so raising the pH in the soil should aid Cr(VI) removal. The first four tests were conducted using a 1.0 VDC/cm voltage gradient, and the fifth experiment was conducted to assess the effect of using a higher 2.0 VDC/cm voltage gradient.

### Chemical Analysis

Following the electrokinetic tests, initial and sectioned soil samples were tested for pH, redox potential, electrical conductivity (EC), and metal concentration (Danda 2001). The anode and cathode solutions were also tested for these chemical parameters. To test the soil, about 5 g of soil was mixed with 10 mL of CaCl<sub>2</sub> in a glass vial, the mixture was then vigorously shaken by hand for a 1 min and the solids were allowed to settle, and the pH, redox potential, and EC of the soil-water mixture were measured. The moisture content of the soil samples was also determined in accordance with *ASTM D 2216*.

For all the experiments, the *USEPA 3050* acid digestion procedure was used on the soil samples to extract the metals (*USEPA 1986*; Chinthamreddy 1999). In addition, the *USEPA 3060A* alkaline digestion procedure was used to extract the Cr(VI) from the soil samples into solution (Chinthamreddy 1999). The solution samples from the acid and alkaline digestion procedures as well as the liquid samples from the batch and electrokinetic experi-

**Table 4.** Contaminant Removal Efficiencies based on Batch Experiments

pH	Chromium removal (%)		Nickel removal %		Cadmium removal (%)	
	0.1 M EDTA	0.2 M EDTA	0.1 M EDTA	0.2 M EDTA	0.1 M EDTA	0.2 M EDTA
2.0	100	96.3	82.6	67.6	84.3	80.8
4.2–4.7	97.2	97.2	81.6	61.7	70.9	78.3
5.0	95.3	96.7	77.1	67.2	74.8	73.6
7.0	95.8	100	78.3	66.0	69.7	68.9
10.0	96.3	100	71.7	62.8	67.8	67.6

Note: EDTA=ethylenediamine tetraacetic acid.

ments were analyzed using an atomic absorption spectrophotometer (AAS). The AAS was used to determine the concentration of chromium, nickel, and cadmium according to USEPA methods 7190, 7520, and 7130, respectively (USEPA 1986). For chromium, the supernatant from the alkaline digestion procedure was analyzed to determine the Cr(VI) concentration in the soil, and then the Cr(III) concentrations were calculated by subtracting the Cr(VI) concentrations from the total chromium concentration determined from the acid digestion procedure.

To assure quality control, duplicate samples were analyzed and a nitrous oxide/acetylene flame was used in AAS analysis. Moreover, the atomic absorption calibration was checked after testing every five samples, and a mass balance analysis was performed for each electrokinetic test. The mass balance differences were generally less than 10% and these differences were mainly attributed to the nonuniform contaminant distribution within the selected soil sample for chemical analysis, as well as to the adsorption of contaminants to the electrodes and/or porous stones.

## Results and Discussion

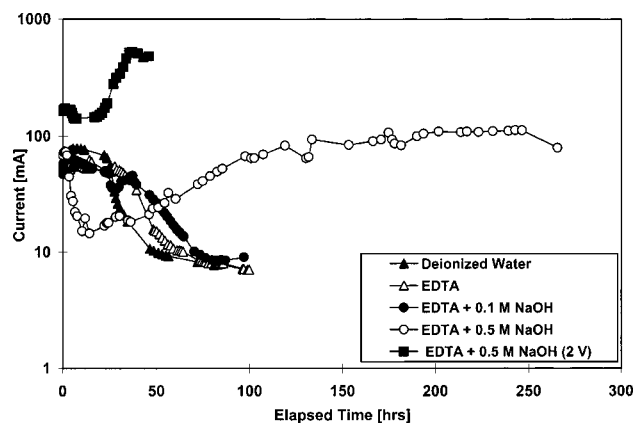
### Batch Experiments

Table 4 presents the results of the batch experiments, and this table shows the removal percentages of Cr(VI), Ni(II), and Cd(II) with the 0.1 and 0.2 M EDTA concentrations at pH values of 2.0, 4.2–4.7, 5.0, 7.0, and 10.0. The removal of chromium was high with both EDTA concentrations and at all the pH values. Chromium removal ranged between  $95 \pm 5\%$ . To assist in estimating the speciation of the metal contaminants in the presence of EDTA in kaolin, a geochemical model (*MINTEQA2*), using a constant capacitance soil adsorption model, was performed (Danda 2001). The modeling results indicated that  $\text{HCrO}_4^-$  and  $\text{Cr}_2\text{O}_7^{2-}$  were the dominant Cr(VI) species at pH values less than about 6.0, and  $\text{NaCrO}_4^-$  and  $\text{CrO}_4^{2-}$  were the dominant species at pH values above 6.0. Overall, based on the batch experiments and geochemical modeling, low Cr(VI) adsorption and high dissolution occurs under a wide range of pH conditions.

The percentage of Ni(II) removed during the batch experiments ranged between 62 and 83%, and, generally, greater removal occurred when the pH was low (between 2.0 and 5.0) (Table 4). The geochemical model indicated that the dominant Ni(II) species should have been Ni-EDTA complexes, such as NiL, NiHL, and/or NiOHL at all the pH values. However, Ni(II) was not completely removed, so some Ni(II) must have adsorbed to the soil or it may have precipitated under high pH conditions. Similar to Ni(II), the percentage of Cd(II) removed ranged from

67 to 84%, and a greater amount of Cd(II) was removed when the pH was low (between 2.0 and 5.0). The geochemical model predicted that the dominant species should have been Cd-EDTA complexes, such as CdL and/or CdHL at all the pH values. Nevertheless, since Cd(II), like Ni(II), was not completely removed, some Cd(II) adsorption may have occurred or a precipitate may have formed under high pH conditions. It should be noted that the removal percentages in the batch experiments were based on the concentrations measured in initially spiked soil samples, and uneven contaminant distribution in these initial soil samples or in the soil used for the individual batch tests could have affected the calculated removal efficiencies.

The results from the batch experiments also show that when the higher 0.2 M EDTA concentration was used, the removal of Cd(II) was greater than that of Ni(II) for all the pH values (Table 4). This indicates that compared to Ni(II), it was thermodynamically favorable for Cd(II) to complex with EDTA at the 0.2 M concentration. Conversely, at the lower 0.1 M EDTA concentration, Ni(II) removal was generally greater than or close to that of Cd(II), so the preferential complexation could have been affected by the EDTA concentration. In addition, for many of the batch experiments, a lower amount of removal occurred when the higher 0.2 M EDTA concentration was used, and this was particularly evident for Ni(II) removal. Overall, the advantage of using a higher EDTA concentration was difficult to discern, but during electrokinetic remediation, it could be beneficial to use the higher 0.2 M concentration to account for EDTA adsorption to the soil and improve EDTA migration into the soil.



**Fig. 2.** Electrical current during electrokinetic treatment

## Electrokinetic Experiments

### Current

Fig. 2 shows the electrical current that occurred during the five electrokinetic tests. As mentioned earlier, the current primarily results from the electromigration of ions through the pore fluid. In the deionized water (baseline) test, the current reached a high value of approximately 78 mA near the start of testing and then began to decrease. It finally appeared to stabilize at around 7 mA after about 100 h. The pattern of decreasing current over time correlates to the findings of other investigators (Eykholt 1992; Grundl and Michalski 1996). The current near the start of testing provides an indication of the amount of ions originally associated with the soil as salt precipitates (or as metal contaminants). As time passes, the current decreases because the mobile ions are constantly electromigrating toward the electrodes, and, as they migrate, these excess ions are neutralized by reacting with the soil, other species in solution, or by reacting with the oppositely charged electrode. The reason for the long-term stabilized current is more difficult to comprehend.  $H^+$  and  $OH^-$  ions are generated at the electrodes due to electrolysis, but when these ions electromigrate toward the oppositely charged electrode, they can meet and react to form water, so their contribution to the current could be minor (Dzenitis 1997). However, the adsorption of these ions to the soil and the slow dissolution of minerals and/or salt precipitates that may result from pH changes could lead to a long-term and steady supply of charge carriers (Eykholt 1992).

As seen in Fig. 2, during around the first 25 h of testing, the current in the experiment using EDTA alone at the cathode had a lower current than the baseline test, but after about 25 h, the current in the baseline test dropped and the EDTA test maintained a slightly higher current for a longer duration. Similarly, after approximately 50 h the current in the experiment employing EDTA at the cathode and 0.1 M NaOH at the anode maintained a higher current for a longer duration than the test using EDTA alone.

It should be mentioned that the introduction of electrolytic solutions with ionic charge carriers, such as Na-EDTA and  $Na^+$  or  $OH^-$  ions, should produce an increased current, but there are many physico-chemical effects that must be considered. Alshwabkeh and Bricka (2000) reported that the effective ionic mobility under a unit electric field depends on a number of factors, such as the charge and size of the particular ion, soil porosity, water content, tortuosity, and pore fluid chemistry. Moreover, generally, ionic transport is enhanced when the electroosmotic flow is concurrent with electromigration, but ionic transport may be hindered when the flow opposes electromigration.

NaOH was introduced at the anode to counteract the production of the low pH (high  $H^+$  concentration) solution generated by the electrolysis reaction. The enhanced supply of  $OH^-$  ions provided by NaOH facilitate the neutralization of  $H^+$  ions by forming water, but the remaining  $Na^+$  ions tend to electromigrate toward the cathode, thereby increasing the current and conductivity in the pore fluid. Thus, it is reasonable that the test using EDTA with 0.1 M NaOH had a slightly higher and more sustained current than the test using EDTA alone (Fig. 2).

The current in the test using EDTA and the 0.5 M NaOH solution dropped from about 70 to 20 mA within the first 25 h or so, but then it gradually increased over the next 100 h and appeared to stabilize near 100 mA after about 150 h. Evidently, as compared to the test with EDTA and 0.1 M NaOH, the current variation in this test was caused by the use of the relatively high 0.5 M NaOH concentration at the anode. As the high  $OH^-$  concentration

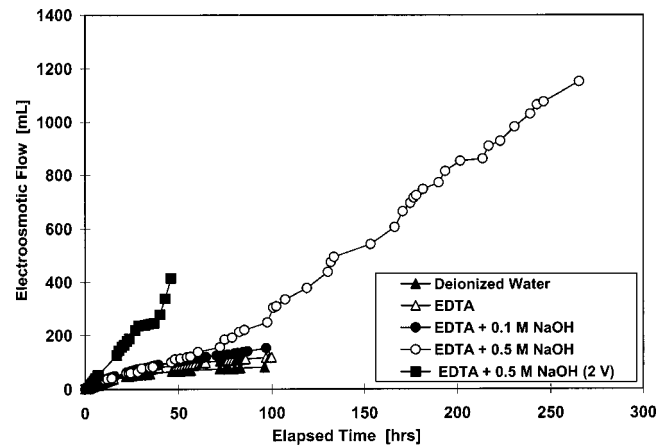


Fig. 3. Cumulative electroosmotic flow during electrokinetic treatment

solution flowed into the soil from the anode due to electroosmosis, the number of mobile cationic species near the start of testing may have been reduced, and, hence, the current was reduced. This is because many cations react with  $OH^-$  ions to form hydroxide precipitates, such as  $Al(OH)_3$ ,  $Ca(OH)_2$ , or  $Cd(OH)_2$ . During this initial time period, the  $Na^+$  ions or other cations that were dissociated may have reacted with anionic species, such as  $CO_3^{2-}$  or  $Cl^-$ , or exchanged with other cations. As time passed, the number of ions passing through the soil due to electromigration and electroosmosis must have increased, and this caused the current to gradually rise. In addition, as time passes, the proportion of  $OH^-$  ions increases at the cathode due to electrolysis, and it is likely that these ions were more mobile and more efficient at carrying charge toward the anode than the EDTA ligand. Wong et al. (1997) observed an increase of current over time, and these researchers attributed the current increase to the increasing  $OH^-$  concentration at the cathode.

In the test employing a 2.0 VDC/cm voltage gradient, the current results exhibited a trend similar to that of the EDTA and the 0.5 M NaOH experiment because the current decreased initially but then increased to values substantially higher than the initial current. Apparently, the same type of reactions as discussed previously occurred due to the relatively high  $OH^-$  concentration, but the higher 2.0 VDC/cm voltage caused the reactions to occur more rapidly and the current values to become much higher. It should be noted that high current values, such as the 500 mA values that resulted near the end of the 2.0 VDC/cm test, might generate large non-uniform pore pressures that could consolidate the soils and adversely affect the remediation process (Eykholt 1997).

### Cumulative Electroosmotic Flow

As mentioned previously, electroosmotic flow is essentially produced by the net migration of ions toward the oppositely charged electrode, and the following Helmholtz-Smoluchowski (HS) equation is often used to estimate the average electroosmotic flow velocity ( $V_{e0}$ ) (Mitchell 1993; Eykholt and Daniel 1994).

$$V_{e0} = - \frac{D\varepsilon_0\zeta}{\eta} E_x \quad (2)$$

According to this equation, the flow velocity is proportional to the electrical gradient ( $E_x$ ), zeta potential ( $\zeta$ ), and dielectric constant ( $D$ ), and it is inversely proportional to the viscosity ( $\eta$ ). The  $\varepsilon_0$

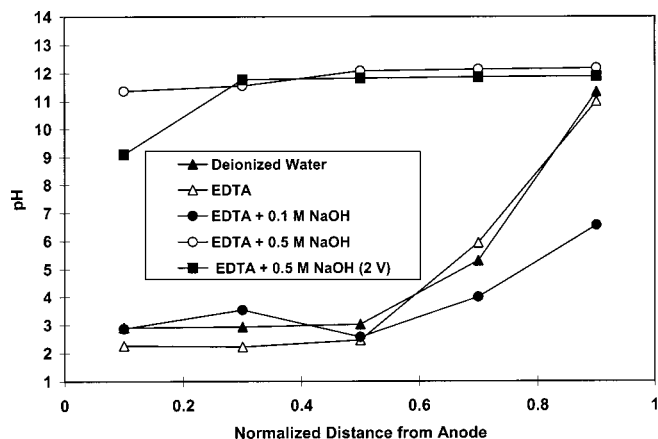


Fig. 4. pH in soil after electrokinetic treatment

term represents the permittivity of vacuum ( $8.854 \times 10^{-12}$  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. When the pH is above the ZPC, the soil particles usually have a negative zeta potential and the electroosmotic flow occurs toward the cathode. Conversely, when the pH is below its ZPC, the soil particle surfaces commonly have a positive zeta potential and the electroosmotic flow occurs toward the anode (reverse electroosmotic flow). However, from the HS equation, it can be seen that in addition to the pH, many other factors might affect the electroosmotic flow. For instance, the surface charge of the soil particles and the zeta potential are affected by changes in the ionic strength, and, other pore fluid properties, such as the dielectric constant and viscosity, also influence the electroosmotic flow. Furthermore, 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).

Fig. 3 shows the cumulative electroosmotic flow that was measured at the cathode. As seen from these results, the cumulative electroosmotic flow measurements somewhat correlated to the electrical current measurements (Fig. 2). This is reasonable because, as mentioned earlier, the electromigration of ions is largely responsible for generating flow. Thus, during the deionized water (baseline) experiment, the current greatly reduced after about 25 h, and this test generated the lowest electroosmotic flow. The current in the experiment with EDTA alone started lower than the baseline test but the current was sustained at a higher level for a slightly longer duration (Fig. 2). As a result, the electroosmotic flow in the EDTA test was slightly higher than in the baseline test (Fig. 3). Similarly, the experiment with EDTA and 0.1 M NaOH generated slightly more flow than the test using EDTA alone.

As observed earlier in Fig. 2, in the experiment using EDTA and 0.5 M NaOH, the current quickly dropped during the first 25 h, and then gradually increased and reached a high stable value. However, as shown in Fig. 3, the electroosmotic flow appears to be gradually increasing during this initial 25 h period. Evidently, during the first 25 h of this experiment, electromigration was occurring and generating electroosmotic flow while the current was reducing. These results corroborate with the earlier hypothesis that the  $\text{Na}^+$  and  $\text{OH}^-$  ions were migrating into the soil from the anode, but the current was reducing because  $\text{OH}^-$  ions were reacting with mobile cations and possibly precipitating, while  $\text{Na}^+$  ions may have been reacting with anions and/or ex-

changing with other cations or adsorbing to the mineral surface. As time passed, the amount of ions being consumed by reactions and adsorption diminished and the mobility of the ions increased, thereby gradually increasing the current and electroosmotic flow.

As observed in Fig. 3, the electroosmotic flow in the EDTA and 0.5 M NaOH experiment increased substantially after approximately 100 h which is nearly the same time that the current reached a comparatively high and stable value of 100 mA (Fig. 2). Finally, as observed earlier in Fig. 2, the experiment employing the high 2.0 VDC/cm voltage gradient with EDTA and 0.5 M NaOH generated high current values that increased to values of around 500 mA near the end of testing. Consequently, as shown in Fig. 3, this experiment produced a much higher cumulative electroosmotic flow compared to the other tests.

### Profile of pH in Soil

Fig. 4 shows the pH values that were measured in the soil sections after the electrokinetic experiments. It is important to recall that the electrolysis reactions produce a low pH solution at the anode and a high pH solution at the cathode. Initially, the pH in the soil for all the experiments was around 7.0, but, as observed in the deionized water (baseline) experiment, the electrolysis reactions cause the pH to vary. In the baseline test, the pH reduced to approximately 3.0 near the anode and it increased to about 11.0 near the cathode. As a result of electromigration and electroosmosis, the low pH (high  $\text{H}^+$  concentration) solution generated at the anode was transported in the soil and moved toward the cathode. Furthermore, the low pH solution migrated faster because the electromigration of  $\text{H}^+$  is concurrent with the electroosmotic flow and the mobility of  $\text{H}^+$  is about 1.76 times that of  $\text{OH}^-$  (Alshabkeh and Bricka 2000).

Fig. 4 indicates that in the baseline, EDTA, and EDTA with 0.1 M NaOH experiments, the low pH (2.0–4.0) solution migrated to approximately the middle of the soil specimen. Conversely, the  $\text{OH}^-$  ions generated by electrolysis at the cathode electromigrate toward the anode, and this caused the pH to rise to around 11.0 in the soil adjacent to the cathode during the baseline and EDTA experiments. Although 0.2 M EDTA is acidic (pH about 4.0), by comparing the baseline and EDTA tests, it can be observed that adding the 0.2 M EDTA solution to the cathode did not cause the pH to become lower near the cathode (Fig. 4). However, compared to the EDTA experiment, the test using EDTA with 0.1 M NaOH resulted in a much lower pH near the cathode. The results indicate that the increased conductivity and electroosmotic flow generated by using the 0.1 M NaOH solution caused the low pH solution to migrate further toward the cathode.

As observed in Fig. 4, in contrast to the EDTA with 0.1 M NaOH experiment, it is clear that the use of the 0.5 M NaOH solution at the anode greatly increased the pH to about 11.0–12.0 throughout the soil. Evidently, this higher concentration was capable of counteracting the  $\text{H}^+$  produced by the electrolysis reaction at the anode, and, as observed earlier, the high pH solution migrated toward the cathode (Fig. 3) due to the high concentration and electroosmosis. These results suggest that the migration of the high pH (high  $\text{OH}^-$  concentration) solution into the soil might have caused  $\text{OH}^-$  to react with cations in the soil, thereby temporarily reducing the current near the start of the test. High pH values, similar to the previously described test, were measured in the soil after conducting the experiment using the high 2.0 VDC-cm voltage gradient, however, the relatively low pH value (9.0) measured adjacent to the anode indicates that an in-

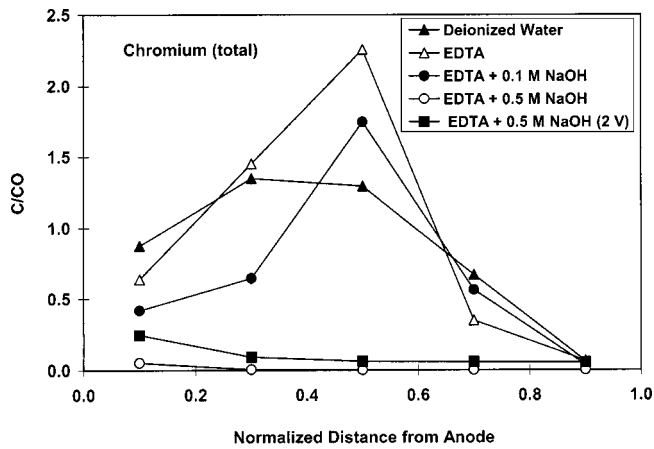


Fig. 5. Total chromium distribution in soil after electrokinetic treatment

creased rate of electrolysis at the anode under this high voltage gradient might cause the pH in the soil to eventually decrease if the test was performed for a longer duration.

#### Residual Contaminant Distribution in Soil

Figs. 5–8 show the chromium (total), Cr(VI), Ni(II), and Cd(II) distribution in the soil specimens, respectively, and Fig. 9 shows the removal efficiencies of these heavy metals. Residual concentrations ( $C$ ) were normalized with initial concentrations ( $C_0$ ) and were plotted in Figs. 5, 7, and 8. Based on the results of the batch experiments, high chromium removal was expected, but, as seen in Fig. 5, the only tests that removed a substantial amount of chromium were the two tests that employed the 0.5 M NaOH solution. Although a sizeable amount of chromium migration occurred during the deionized water (baseline), EDTA, and EDTA with 0.1 M NaOH experiments, very little chromium was removed into the cathode or anode reservoirs. Fig. 6 illustrates that most of the chromium remaining in the soil existed in the Cr(VI) form, which, as stated earlier, primarily exists as anionic complexes, such as  $\text{CrO}_4^{2-}$  at high pH (greater than 6.0) and as  $\text{Cr}_2\text{O}_7^{2-}$  or  $\text{HCrO}_4^-$  at low pH (less than 6.0). Therefore, Cr(VI) species electromigrate toward the anode, which opposes the direction of electroosmotic flow. Under low pH conditions, Cr(VI) species may adsorb to the soil, and this is apparently what happened

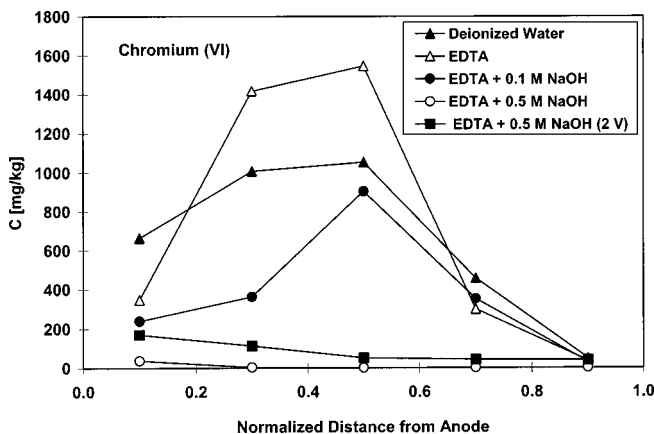


Fig. 6. Hexavalent chromium [Cr(VI)] distribution in soil after electrokinetic treatment

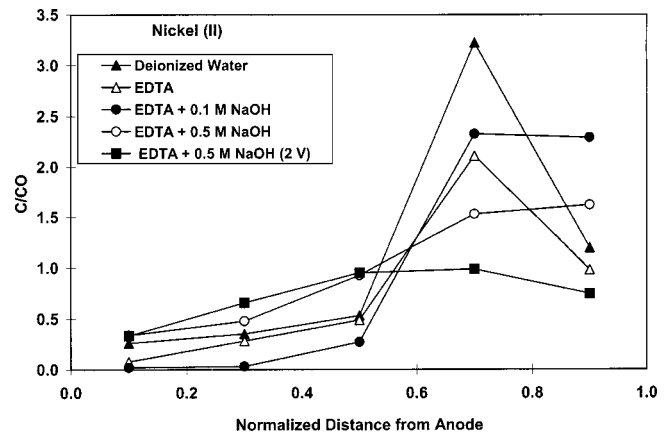


Fig. 7. Nickel distribution in soil after electrokinetic treatment

when the Cr(VI) anionic complexes entered the low pH region during the baseline, EDTA, and EDTA with 0.1 M NaOH tests. Conversely, the results indicate that the high pH conditions that existed in the soil during the tests employing the 0.5 M NaOH solution (Fig. 4) facilitated high (85–95%) chromium removal (Fig. 9). It should be noted that Cr(VI) was removed at the anode reservoir, indicating that it electromigrated in a direction opposite to the electroosmotic flow. This strongly suggests that electromigration was the dominant electrokinetic transport process for the removal of Cr(VI) species.

According to the geochemical model (*MINTEQA2*), when EDTA is not present, Ni(II) exists as the  $\text{Ni}^{2+}$  cation when the pH is less than about 6.0, and it precipitates as  $\text{Ni}(\text{OH})_2$  when the pH becomes greater than around 8.0 (Danda 2001). As seen in Fig. 7, for the baseline test, the normalized Ni(II) concentration was relatively low in the soil region where the pH was low, but the concentration increased substantially near the cathode where the pH increased. Evidently, during the baseline test, Ni(II) dissolution occurred under the low pH conditions near the anode, and then  $\text{Ni}^{2+}$  ions electromigrated toward the cathode. However, when the  $\text{Ni}^{2+}$  ions entered the high pH conditions near the cathode (Fig. 4), they adsorbed or precipitated as  $\text{Ni}(\text{OH})_2$ . Thus, although Ni(II) migrated toward the cathode and the concentration reduced significantly through the half of the soil specimen nearest to the anode, the amount of Ni(II) that was completely removed from the soil in the baseline test was negligible.

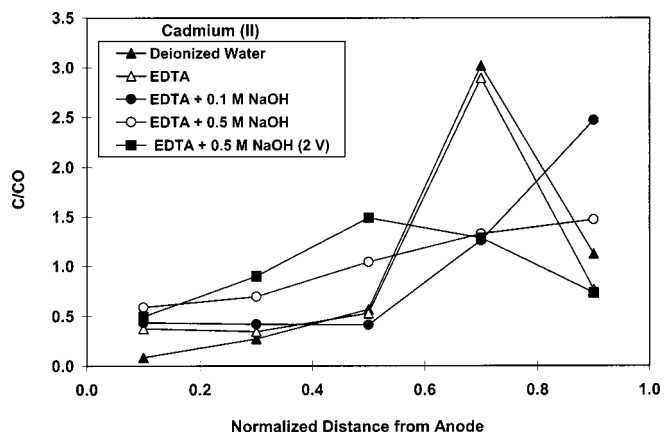


Fig. 8. Cadmium distribution in soil after electrokinetic treatment

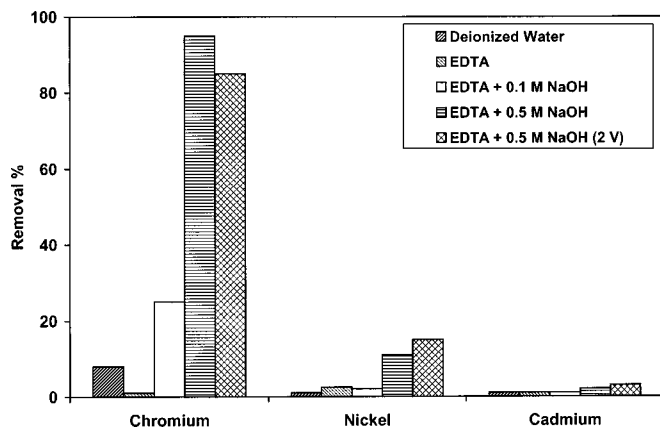


Fig. 9. Removal percentages of chromium, nickel, and cadmium

As observed during the batch experiments (Table 4), the use of the EDTA solution resulted in fairly high Ni(II) removal efficiency (60–80%), but Fig. 7 shows that only a small amount of Ni(II) was removed during the electrokinetic experiments that used EDTA or EDTA with 0.1 M NaOH. The sorption of Ni(II) ions or the precipitation of Ni(OH)<sub>2</sub> near the cathode and the low amount of Ni(II) removal in these tests indicates that EDTA complexation with Ni(II) in these tests was limited. As mentioned earlier, EDTA is a large ligand relative to an anion such as OH<sup>-</sup>, so it has a comparatively low ionic mobility. Moreover, the migration of EDTA anionic complexes was toward the anode, opposite to the direction of electroosmotic flow. Therefore, the amount of EDTA electromigration into the soil from the cathode may have been insufficient during these two electrokinetic experiments, and, since EDTA electromigration into the soil was hindered, Ni–EDTA complexation was inadequate.

A minor amount of Ni(II) was removed during the two tests that used the EDTA and 0.5 M NaOH flushing solutions, and Fig. 7 shows that a substantial amount of Ni(II) migrated toward the cathode during these tests. This suggests that during the first few hours of testing a low pH solution generated by the electrolysis reaction at the anode flowed into the soil and was transported toward the cathode, and this solution facilitated Ni<sup>2+</sup> migration. It is likely that this initial flow of low pH solution was also responsible for the migration of a minor amount (11–15%) of the Ni(II) into the cathode reservoir (Fig. 9). Subsequently, the high pH (about 12.0) 0.5 M NaOH solution raised the pH at the anode by counteracting the electrolysis reaction, and this high pH solution migrated into the soil toward the cathode due to the high concentration and electroosmotic flow. The high pH solution causes cations, such as Ni<sup>2+</sup> to adsorb or precipitate as hydroxides, such as Ni(OH)<sub>2</sub>, and this would produce a reduction in current.

According to the batch experiments (Table 4), Cd(II) removal was expected to be high (67–80%), but, although a substantial amount of Cd(II) migration occurred, similar to what was observed with Ni(II), most of the Cd(II) remained within the soil specimen (Fig. 8). Slight differences can be observed between the distribution of Ni(II) and Cd(II) in Figs. 7 and 8, but, generally, the migration of these heavy metals was comparable. Thus, the migration of Cd(II) can be attributed to the same electrokinetic transport mechanisms that were used to describe the migration of Ni(II).

The results of this investigation are fairly consistent with other studies that used electrokinetics with EDTA conditioning at the cathode to remediate lead-contaminated soils. In EDTA-enhanced

electrokinetic tests conducted by Reed et al. (1995), the majority of the lead migrated toward the cathode in the same direction as the electroosmotic flow. These investigators concluded that it was unlikely that EDTA formed complexes with lead, and the lead probably migrated due to the low pH conditions generated by electrolysis at the anode. Apparently, like the results of the present investigation, these researchers also found that EDTA electromigration toward the anode was hindered when it opposed the electroosmotic flow. Conversely, Yeung et al. (1996) attempted to remediate a lead-contaminated Milwhite kaolinite, and the majority of the lead migrated toward the anode. However, in this investigation the electroosmotic flow was in the reverse direction, toward the anode, apparently due to the presence of iron and titanium oxide impurities. Thus, these other studies reaffirm the hypothesis that the large size and low mobility of the EDTA molecules limit EDTA electromigration toward the anode when it opposes the direction of electroosmotic flow.

## Summary and Conclusions

When electrokinetics was used with de-ionized water as a flushing solution, Cr(VI) (anionic species) migrated toward the anode, whereas Ni(II) and Cd(II) (cationic species) migrated toward the cathode. Although substantial heavy metal migration occurred, the amount of contaminant removal was minimal, largely due to the changes in pH caused by the electrolysis reactions at the anode and cathode. Cr(VI) species tend to adsorb to the soil under the low pH conditions that exist near the anode, and Ni(II) and Cd(II) tend to precipitate under the high pH conditions that exist near the cathode.

Batch experiments revealed that high heavy metal removal was possible using either a 0.1 or 0.2 M EDTA concentration under a wide range of pH conditions. However, the results of the electrokinetic experiment using EDTA at the cathode showed low heavy metal removal efficiency. Since Cr(VI) species tend to adsorb to the soil under low pH conditions, it was hypothesized that Cr(VI) removal might be improved by increasing the pH in the soil near the anode. Moreover, raising the pH might prevent Ni-EDTA and Cd-EDTA complexes from dissociating with these heavy metals and preferentially complexing with H<sup>+</sup> under low pH conditions near the anode. Consequently, experiments were conducted using different concentrations (0.1 and 0.5 M) of NaOH at the anode to increase the pH through the soil and improve contaminant removal.

The results of the NaOH experiments indicated that a relatively high 0.5 M NaOH concentration was necessary to counteract the electrolysis reaction and raise the pH across the soil specimen and, under these high pH conditions, a significant (95%) amount of Cr(VI) removal was achieved at the anode. This removal was attributed to electromigration since the Cr(VI) species migrated in a direction opposite to the electroosmotic flow. In addition, before the high pH solution could counteract the electrolysis reaction at the anode and raise the pH, it appears that low pH solution moved through the soil toward the cathode. This initial flow of low pH solution resulted in a substantial amount of Ni(II) and Cd(II) migration toward the cathode, but the remedial efficiency of these heavy metals was low due to adsorption or precipitation near the cathode. Furthermore, very little Ni(II) or Cd(II) migration occurred toward the anode, so EDTA complexation with the heavy metals was deemed to be inadequate. Ethylenediamine tetraacetic acid electromigration was most likely limited due to its relatively large size and low mobility, and the

direction of EDTA migration opposed the direction of electroosmotic flow. Finally, an electrokinetic experiment was conducted using the same 0.5 M NaOH and 0.2 M EDTA flushing solutions at the anode and cathode, respectively, along with a comparatively high 2.0 VDC/cm voltage gradient. Like the identical test performed with the 1.0 VDC/cm voltage gradient, the high voltage test achieved high (85%) Cr(VI) removal at the anode, but, as in the lower voltage test, Ni(II) and Cd(II) removal was low.

This study shows that there are many complicating factors that affect the EDTA-enhanced electrokinetic remediation of multiple heavy metals in clayey soils. First, it is important to understand the chemistry of each particular metal contaminant and its behavior under different subsurface conditions. Additional considerations are the pH variation through the soil that is caused by the electrolysis reactions, the low mobility of EDTA, and the opposing direction of electroosmotic flow. Adjusting the pH through the soil by using a high pH solution to counteract the electrolysis reaction at the anode was found to be advantageous for Cr(VI) removal, but it was evident that the increased electroosmotic flow hindered EDTA electromigration toward the anode. In addition, the use of a relatively high 2.0 VDC/cm voltage gradient was determined to cause a high current that increased the electrolysis reaction. Overall, this study found that many factors influence heavy metal removal during EDTA-enhanced electrokinetic remediation and all of these factors must be carefully evaluated and researched further to enhance contaminant removal.

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