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Technical Challenges to In-situ Remediation of Polluted Sites

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Abstract Throughout the world, subsurface contamination has become a widespread and pervasive problem. Toxic chemicals such as heavy metals and organic compounds are commonly used in a myriad of industries. However, largely through inadvertent or accidental release, these chemicals are presently polluting sites across the United States. In order to protect public health and the environment, further pollution must be prevented and sites with existing contamination urgently need remediation. Unfortunately, remediating subsurface contamination has proved to be a daunting task. Heavy metals and organic compounds often coexist and their distribution within the subsurface is highly dependent on particle and macro-scale heterogeneities. Vast resources have been invested to develop efficient remediation technologies, yet very few of these technologies have been successful. In-situ remediation is often preferred due to minimal site disturbance, safety, simplicity, and cost-effectiveness. The effectiveness of in-situ remediation technologies depends largely on the contaminant chemistry and subsurface heterogeneities (including particle-scale heterogeneities such as fine-grained soils, soils with reactive minerals, and/or soils rich in organic matter as well as macro-scale heterogeneities such as irregular soil layers and/or lenses). Under such heterogeneous conditions, integrated electrokinetic remediation technology has great potential. As a safe and economical remedial option for so many contaminated sites, the application of integrated electrokinetic remediation offers enormous public health, environmental, and financial benefits.

Keywords Electrokinetic remediation · Soils · Pollution · Heavy metals · Organic compounds

1 Introduction

Polluted sites pose a serious hazard to public health and the environment. The United States Environmental Protection Agency (USEPA) estimates that over 217,000 sites require urgent cleanup at an estimated cost of over $187 billion (USEPA 1997). Soil and groundwater contamination has been a major problem at the sites in question. The contaminants encountered at these sites include metals (such as lead, chromium, nickel, strontium, and uranium), volatile organic compounds (such as benzene, toluene, and trichloroethylene), and semi-volatile organic compounds (such as polycyclic aromatic hydrocarbons (PAHs) and polychlorinated biphenyls (PCBs)). Organic and metal contaminants are found to coexist at many sites.

Recently, environmental professionals have focused on risk-based approaches to remediating

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polluted sites. Risk assessment includes detailed site characterization, human and ecological risk quantification, and selection of remedial goals (Sharma and Reddy 2004). The end-use of the site is also taken into consideration in the risk assessment process. If the risk posed by the contamination is unacceptable, a remedial action must be chosen and implemented to achieve the remedial goals in an efficient and cost-effective manner. Remediation of polluted sites can help preserve green lands (pristine land) from pollution caused by industrial development as well as provide opportunities for economic growth.

Several technologies have been developed to remediate contaminated sites (Sharma and Reddy 2004). These technologies can be grouped as either ex-situ or in-situ technologies. Ex-situ remediation technologies involve removing the contaminated soil and/or groundwater from the subsurface and treating it on-site or off-site. Conversely, in-situ remediation technologies involve treating the contaminated soil and/or groundwater in place without removing it from the subsurface. Often, in-situ remediation technologies are preferred because they result in minimal site disturbance and expose workers and the surrounding public to the lowest amount of contaminants. In addition, in-situ technologies are often less costly due to simpler procedures. Common in-situ soil remediation technologies include soil vapor extraction, soil flushing, solidification and stabilization, thermal desorption, vitrification, bioremediation, and phytoremediation. Common in-situ groundwater remediation technologies include pump and treat, air/ozone sparging, flushing, permeable reactive barriers, immobilization, chemical oxidation, and bioremediation. All of these remediation technologies are based on physico-chemical, thermal, or biological processes that aim to remove the contaminants from soils and groundwater or to immobilize and/or detoxify the contaminants within the soils and groundwater.

This article summarizes the common technical challenges in remediating contaminated sites in-situ and subsequently provides a description of integrated electrokinetic remediation, an alternative technology that has the potential to surmount such obstacles.

2 Technical Challenges

The USEPA has documented the inadequate performance of various remediation technologies implemented at numerous polluted sites (USEPA 2000, 2006). Poor performance may be attributed to the following technical difficulties: (1) clayey soils are difficult to remediate because of low permeability and complex composition (mineralogy and organic content); (2) many common technologies fail under heterogeneous subsurface conditions (e.g., clay lenses within sand formation); (3) hydrophobic organic contaminants such as PAHs and PCBs are difficult to treat; (4) very few technologies are available to cleanup sites contaminated with heavy metals; and (5) very few technologies exist that can remediate sites contaminated by mixed contaminants (e.g., organic compounds combined with heavy metals and/or radionuclides). There is an urgent need to develop new cost-effective technologies that can overcome these challenges. In-situ electrokinetic remediation technology has great potential to be one of these technologies (Acar et al. 1995; Page and Page 2002).

An electrokinetic remediation system for the in-situ treatment of contaminated sites consists of drilling wells or drains in which electrodes are installed and then applying a very low direct current electric potential. Pumping and conditioning systems may be needed at the electrodes in some situations. A schematic of in-situ electrokinetic remediation system is shown in Fig. 1. Similarly, electrokinetic treatment may be accomplished ex-situ by using specially designed above-ground reactors. Generally, the contaminants accumulated at the electrodes are removed by either adsorption onto the electrodes or withdrawal followed by treatment. Electrokinetic remediation offers the following advantages as compared to conventional remediation methods: (1) simplicity—minimal equipment requirements; (2) safety—neither personnel nor the public in the vicinity are exposed to contaminants; (3) wide range of contaminated media—can be used for soils, sludges, sediments, and groundwater (particularly well suited for low-permeability clays and heterogeneous soil deposits within the vadose zone where conventional remedial methods have proven to be ineffective or expensive); (4) wide range of contaminants—can be used for metals, organic compounds, radionuclides, or combinations of these contaminants; (5) flexibility—can be used as an in-situ or ex-situ remediation system and is easily integrated with other remediation technologies such as

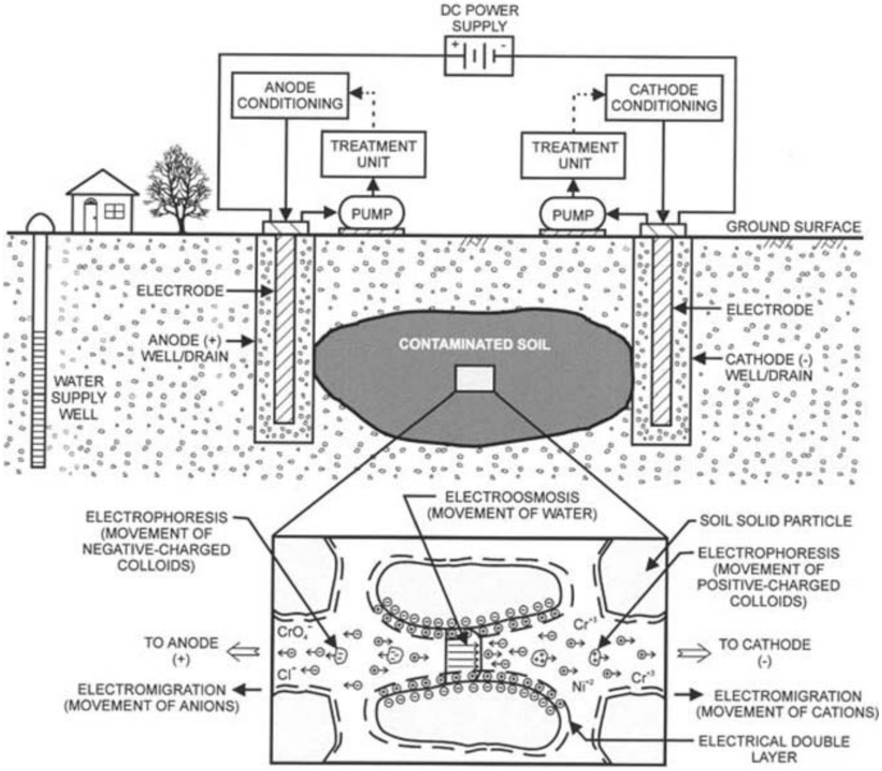


Fig. 1 Schematic of in-situ electrokinetic remediation system

bioremediation; and (6) cost-effectiveness—requires low electrical energy leading to lower overall cost, with costs ranging from $20 to $225 per cubic yard depending on site-specific conditions.

The successful implementation of electrokinetic remediation requires a thorough understanding of the transport mechanisms and physico-chemical processes that affect the transport and fate of contaminants under an induced electric potential (Acar et al. 1995; Alshawabkeh et al. 1999). The major contaminant transport mechanisms under an induced electric potential are: (1) Electroosmosis—bulk movement of pore fluid through the electrical double layer in clayey soils, generally occurring from anode to cathode; (2) electromigration—transport of ions and ion complexes within the pore fluid towards oppositely charged electrodes; (3) electrophoresis—transport of charged colloids, micelles, bacterial cells, etc. within the pore fluid towards oppositely charged electrodes; and (4) diffusion—transport of chemicals due to concentration gradients. Electroosmosis is the major transport process for non-polar organic compounds, while electromigration is the dominant transport process for ionic chemical species. These two processes govern the overall contaminant migration in compact soil systems; and the role of the other two processes, electrophoresis and diffusion, is often negligible. However, electrophoresis may be a significant contaminant transport process in sludge and sediments.

Research conducted to date shows that the electrochemical processes are quite complex and are influenced by the site-specific geochemical environment. As a result of the induced electric potential, electrolysis of water occurs at the electrodes. The electrolysis reactions generate H^+ ions and oxygen gas at anode and OH^- ions and hydrogen gas at cathode. The gases may be allowed to emit into the atmosphere, while the H^+ ions migrate towards cathode and OH^- ions migrate towards anode. Depending on the extent of the migration of H^+ and OH^- ions, pH changes occur across the soil. Generally, low pH (acidic) conditions exist near anode and high pH (basic) conditions exist near cathode (Acar et al. 1995). The pH changes in the soil will affect the geochemical processes, namely adsorption and desorption, precipitation and dissolution, and oxidation and reduction.

The lower soil pH near anode causes desorption and solubilization of cationic (negatively charged) metals, such as lead, nickel and cadmium, enhancing their electromigration towards cathode. However, the higher pH near cathode causes these metals to adsorb and/or precipitate, hindering electromigration and removal at cathode. The change in pH also affects the surface charge of soil particles. If the pH is less than the point of zero charge (PZC), the soil surfaces are positively charged. However, if the pH is greater than the PZC, the soil surfaces will be negatively charged. PZC is the pH at which the net charge on the particle surfaces is zero. The changes in surface charge will affect the diffuse double layer, consequently affecting the electroosmotic flow. The changes in surface charge also affect adsorption of the contaminants to the soil surfaces. Thus, changes in surface charge will affect the migration and removal of both organic and metal contaminants. Therefore, the effects of pH and other geochemical (redox potential, electrolyte concentration, etc.) changes under applied electric potential on the site-specific soil and contaminant conditions and remediation should be assessed.

For electrokinetic remediation to be feasible, contaminants must be desorbed and/or solubilized in the soil. H^+ transport causes desorption/solubilization of cationic metals, while OH^- transport causes desorption of anionic metals. Electromigration of cations and anions occurs towards cathode and anode, respectively. Electroosmotic advection also causes ions and non-polar organic contaminants to transport towards the electrodes. The contaminants are then captured in the electrolyte at the electrodes and treated above-ground using common wastewater treatment technologies.

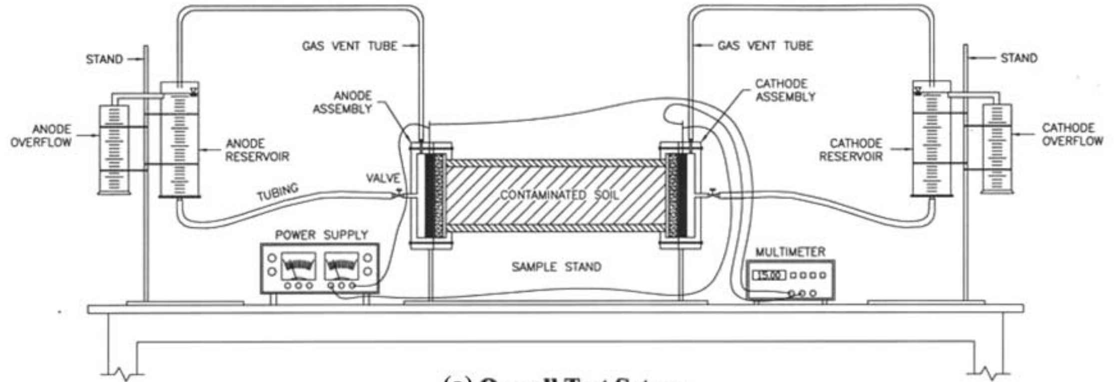
3 Enhanced Electrokinetic Remediation

Several researchers have investigated potential applicability of electrokinetic remediation of soils contaminated with heavy metals and organic compounds (Acar et al. 1995; Page and Page 2002; Sawada et al. 2004; Amrate et al. 2005; Ribeiro et al. 2005; Deng and Jennings 2006; Niqui-Arroyo and Ortega-Calvo 2007; Isosaari et al. 2007). A comprehensive electrokinetic research program has been in place at the University of Illinois at Chicago since 1993. The main objectives of this research program are to: (1) investigate geochemistry during electrokinetic remediation in different soil composition and contaminant environments, and (2) investigate methods to engineer geochemistry to favor enhanced contaminant remediation. The results of this research have provided comprehensive fundamental knowledge necessary to develop electrokinetic remediation as a practical soil and groundwater remediation technology. The research approach includes conducting bench-scale experiments to investigate process fundamentals and optimal operational parameters, and mathematical modeling to serve as a screening and optimization tool. Figure 2 shows a typical bench-scale electrokinetic test setup (Reddy and Parupudi 1997; Reddy et al. 1997). It demonstrates that common technical challenges can be overcome by the implementation of enhanced electrokinetic remediation systems.

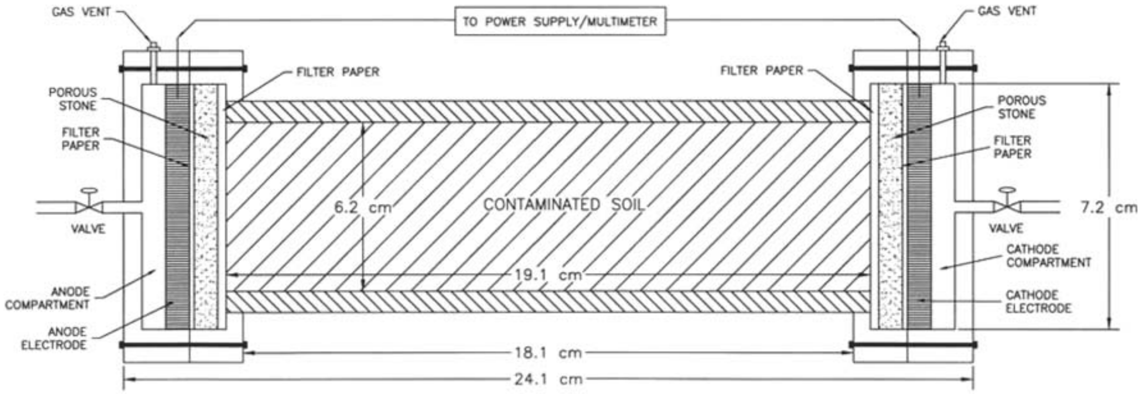
3.1 Removal of Heavy Metals

Bench-scale experiments have provided valuable information on the geochemistry and transport of heavy metals under applied electric potential (Reddy et al. 2001; Al-Hamdan and Reddy 2005). Experiments have been conducted on widely varying clayey soils, including kaolin and glacial till, contaminated with heavy metals such as chromium (Cr), nickel (Ni), cadmium (Cd), and mercury (Hg). Typical results shown in Fig. 3a demonstrate that cationic metals migrate towards cathode, but their migration is retarded by high pH near cathode. In contrast, as shown in Fig. 3b, anionic metals migrate towards anode, but their migration is retarded by low pH near anode. Electromigration is the dominant transport process for heavy metals.

The different compositions of soils lead to various contaminant migration behavior. For example, glacial till soil possesses a high acid buffering capacity due to its high carbonate content and the soil remains alkaline even after the application of electric potential. The high soil pH hinders the migration of cationic metals and enhances the migration of anionic metals (Fig. 3c, d). The presence of multiple contaminants is also shown to hinder migration and removal of contaminants (Reddy and Parupudi 1997). A detailed understanding of speciation and distribution of contaminants before and after electrokinetic remediation has been developed by various geochemical analyses



(a) Overall Test Set-up



(b) Cell Details

Fig. 2 Schematic of typical bench-scale electrokinetic test setup

including XRD, TEM, and sequential chemical extractions as well as geochemical modeling (Reddy et al. 2001; Al-Hamdan and Reddy 2005).

It has been demonstrated that using water as an electrolyte results in nominal removal of heavy metals from the soils. As a result, different strategies have been investigated to enhance removal efficiency. These strategies include increasing treatment duration, increasing electric potential gradient, applying electric potential in different modes (e.g., continuous, periodic, etc.), using cation/anion exchange membranes in the electrodes, circulating electrolytes, and using enhancement (electrode conditioning) solutions such as organic acids (e.g., acetic acid and citric acid) and chelating agents (e.g., EDTA, DTPA). These enhancement strategies resulted in removal of chromium, nickel, and cadmium to levels as high as 98% (Reddy and Chinthamreddy 2003, 2004; Reddy et al. 2004).

Reddy et al. (2003a, b) investigated the use of an iodide-enhanced solution at the cathode during electrokinetic treatment to optimize the removal of mercury from soils. The experimental program consisted of testing two types of clayey soils, kaolin and glacial till, which were initially spiked with 500 mg/kg of Hg(II). Experiments were conducted on each soil type at two voltage gradients, 1.0 or 1.5 VDC/cm, to evaluate the effect of the voltage gradient when employing a 0.1 M KI solution. Additional experiments were performed on each soil type to assess the effect of using a higher iodide concentration of 0.5 M KI when using a 1.5 VDC/cm voltage gradient. The tests conducted on the kaolin soil showed that when the 0.1 M KI concentration was employed with the 1.0 VDC/cm voltage gradient, approximately 97% of the mercury was removed, leaving a residual concentration of 16 mg/kg in the soil after treatment.

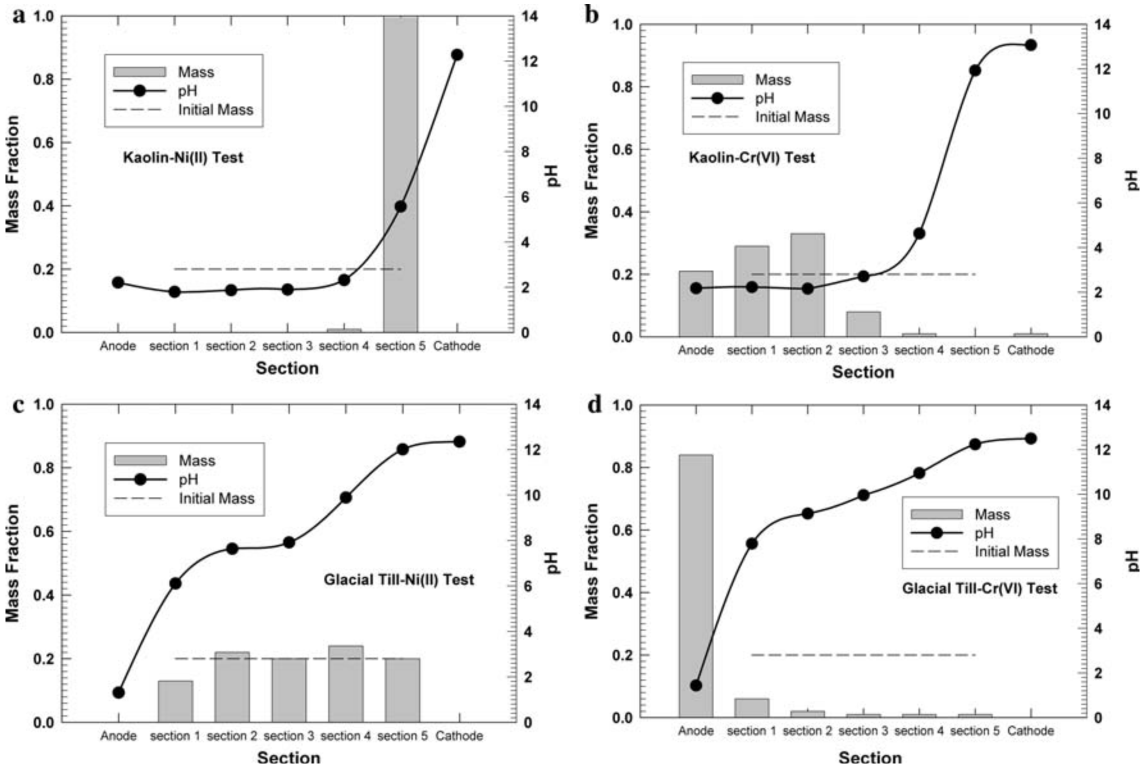


Fig. 3 (**a**) Migration and removal of Ni(II) in kaolin due to electrokinetic treatment. (**b**) Migration and removal of Cr(VI) in kaolin due to electrokinetic treatment. (**c**) Migration and removal of Ni(II) in glacial till due to electrokinetic treatment. (**d**) Migration and removal of Cr(VI) in glacial till due to electrokinetic treatment

The tests conducted on glacial till indicated that it is beneficial to use the higher 0.5 M KI concentration and the higher 1.5 VDC/cm voltage gradient to enhance mercury removal, because, under these conditions, a maximum of 77% of the mercury was removed from the glacial till, leaving a residual concentration of 116 mg/kg in soil after electrokinetic treatment. Compared to kaolin, the lower mercury removal from the glacial till soil is attributed to the more complicated soil composition such as the presence of carbonates and organic matter, which caused Hg(II) to adsorb to the soil and/or exist as an immobile chemical species.

3.2 Removal of Hydrophobic Organic Compounds

Hydrophobic organic compounds (HOCs) are difficult to remove from soils due to their low solubility and strong adsorption to soil surfaces and organic matter in low-permeability clayey soils. Electrokinetically enhanced in-situ flushing using solubilizing agents, such as surfactants, cosolvents, and cyclodextrins, has the potential to remove HOCs from low-permeability clay soils. Previous research has shown that the applied electric potential produces complex physical, chemical, and electrochemical changes within clay soils that affect mass transfer and overall removal efficiency (Li et al. 2000; Reddy and Saichek 2003; Saichek and Reddy 2003; Khodadoust et al. 2006).

Recently, it has been shown that the HOC removal is greatly enhanced by using surfactant-enhanced pulsed electrokinetic treatment (Fig. 4). Pulsed electrokinetics consists of applying a periodic voltage application according to a cycle of 5 days of continuous treatment followed by 2 days of "down time," when the voltage is not applied (Reddy and Saichek 2004). The periodic voltage effects were evaluated by performing four different bench-scale electrokinetic tests with the voltage gradient applied continuously or periodically, under relatively low voltage (1.0 VDC/cm) and high anode buffering (0.1 M NaOH) as well as high voltage (2.0 VDC/cm) and low anode buffering (0.01 M NaOH) conditions. For all the tests, kaolin soil was used as a representative clay soil and was spiked with phenanthrene, a representative HOC, with a target concentration of 500 mg/kg. A non-ionic polyoxyethylene surfactant, Igepal CA 720, was used as the flushing solution in all the tests.

The results of these experiments show that considerable contaminant removal can be achieved by employing a high, 2.0 VDC/cm, voltage gradient along with a periodic mode of voltage application (Fig. 4a). The increased removal was attributed to increased phenanthrene solubilization and mass transfer due to the reduced flow of the bulk solution during the down time as well as to the pulsed electroosmotic flow that improved flushing action (Fig. 4b). Overall, such studies have shown that electrokinetic remediation is a viable technique to remove HOCs from soils.

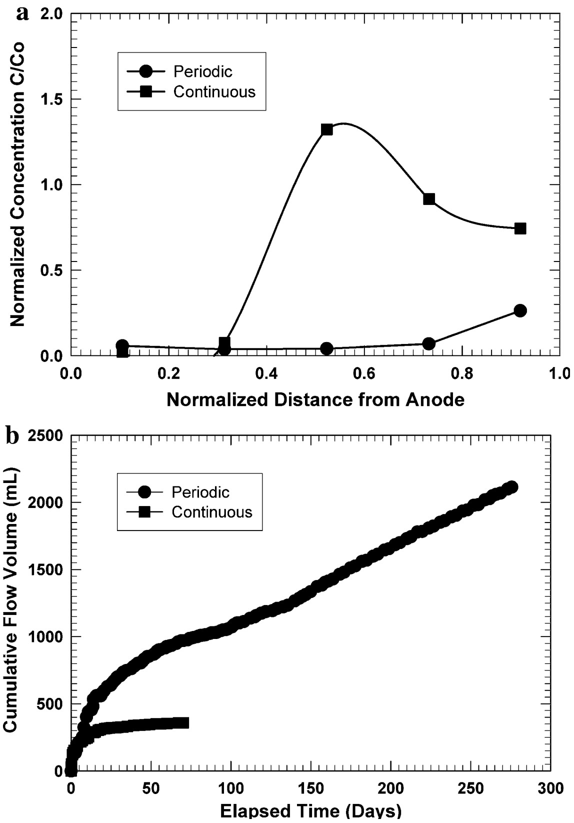


Fig. 4 (**a**) Migration and removal of phenanthrene in kaolin during pulsed (periodic) electrokinetic treatment. (**b**) Electroosmotic flow in kaolin during pulsed (periodic) electrokinetic treatment

3.3 Remediation of Heterogeneous Subsurface

Generally, subsurface conditions are heterogeneous in nature consisting of clay layers and sand layers and/or clay layers interbedded in sand formations. Common remediation techniques based on hydraulic flushing can only remediate sand formations and the contamination associated with clay layers/lenses cannot be remediated. Saichek and Reddy (2005) demonstrated the applicability of electrokinetic remediation technology to treat contaminated soils under soil heterogeneities such as layers, lenses, and mixtures of different soils. Specifically, this study evaluated surfactant-enhanced electrokinetic remediation of PAHs under heterogeneous soil conditions. A series of bench-scale experiments was conducted using two soils (sand and kaolin) spiked with a representative PAH compound (phenanthrene) in a two-dimensional electrokinetic test apparatus under various layered, lens, or mixed soil configurations (Fig. 5). In addition, the homogeneous sand and kaolin soils were each tested alone for comparison purposes. All experiments employed the same non-ionic surfactant (5% Igepal CA-720) flushing solution and a low (0.05) hydraulic gradient.

The results showed that surfactant flushing under the low hydraulic gradient alone was sufficient for complete removal of the contaminant from the homogeneous sand profile (Fig. 5a), whereas the electroosmotic flow generated by the application of a DC 2.0 V/cm electric potential in a periodic mode considerably enhanced the removal efficiency for the homogeneous and heterogeneous soil profiles containing kaolin (Fig. 5b, c and d). The voltage gradient varied spatially and temporally through the soil profiles and affected the electroosmotic flow and contaminant removal (Saichek and Reddy 2005).

3.4 Remediation of Mixed Contaminants

Common remediation technologies are applicable for either heavy metals or organic contaminants. However, mixed contaminants, a combination of heavy metals and organic contaminants, are often encountered at sites (e.g., manufactured gas plant sites). Electrokinetic remediation can be integrated with conventional remediation technologies at sites where mixed contaminants are found. Electrokinetic remediation can induce substantial uniform electroosmotic

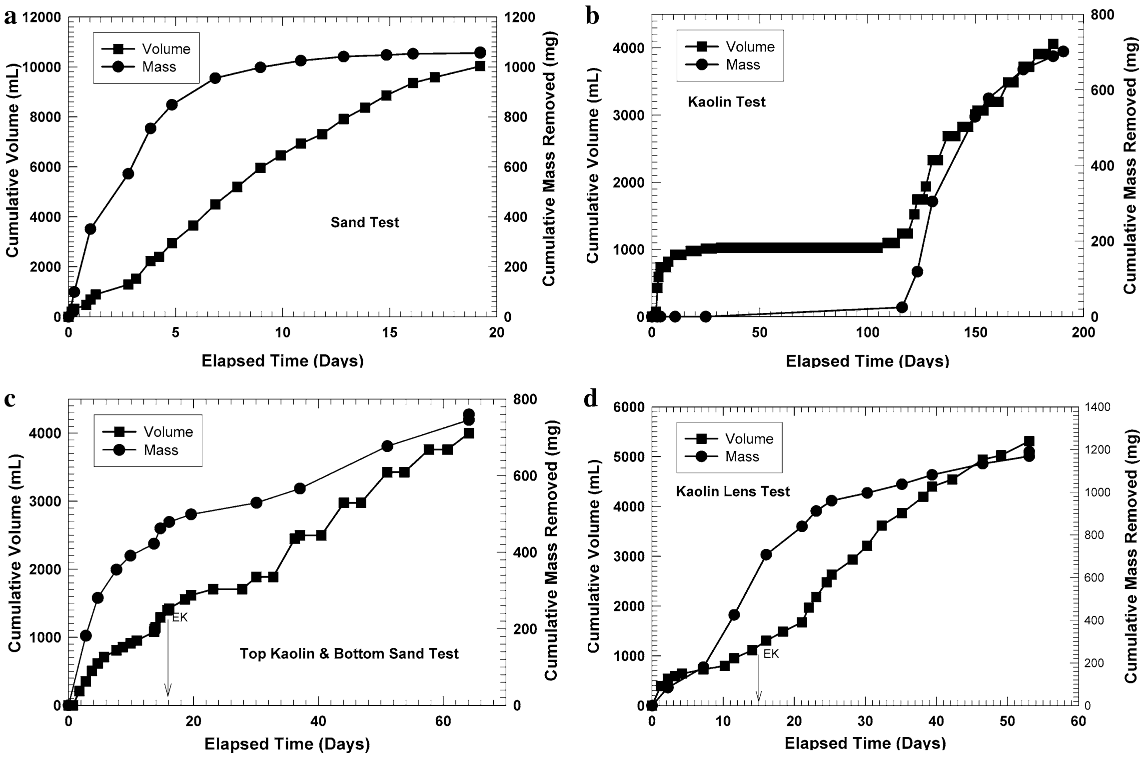


Fig. 5 (**a**) Flow and mass of phenanthrene removal from sand during surfactant flushing. (**b**) Flow and mass of phenanthrene removal from kaolin during electrokinetically enhanced surfactant flushing. (**c**) Flow and mass of phenanthrene removal from soil profile with top clay layer and bottom sand layer during electrokinetically enhanced surfactant flushing. (**d**) Flow and mass of phenanthrene removal from sand layer with interbedded clay lense during electrokinetically enhanced surfactant flushing

flow through low-permeability and heterogeneous soils. It can also remove both metals and organic contaminants by electromigration, electroosmosis, and electrophoresis. Electrokinetic remediation can be integrated with common technologies such as pump and treat, soil flushing, permeable reactive barriers, chemical stabilization/chemical oxidation, in-situ bioremediation, and soil heating. For example, Maturi and Reddy (Reddy and Maturi 2005; Maturi and Reddy 2006) evaluated the simultaneous removal of co-existing heavy metals and PAHs from soils having low permeability using electrokinetics. Nickel and phenanthrene were used as representative heavy metal and organic contaminants, respectively. Different flushing solutions were evaluated to enhance solubilization and transport of hydrophobic phenanthrene (Fig. 6). The experiment with surfactant as a flushing solution resulted in complete removal of phenanthrene.

Other flushing solutions did not result in complete removal of phenanthrene. In the experiment with cyclodextrin, approximately one pore volume of flushing resulted in approximately 50% phenanthrene removal from the soil near anode. However, further migration was retarded because of the reduced electroosmotic flow. In the experiment with a cosolvent as a flushing solution, though the electroosmotic flow was high, lower solubility of phenanthrene in the cosolvent due to the insufficient concentration caused the low removal of phenanthrene. Nickel was found to migrate towards cathode and most of it accumulated within the soil close to cathode due to the high pH conditions generated by electrolysis reaction in all the tests excluding the test with the cosolvent (Fig. 6). When the cosolvent was employed, nickel precipitated throughout the soil because of the high pH of the cosolvent solution. It was concluded that solubilization of the contaminants and control of soil pH are the critical factors that contribute to sustained electroosmotic flow and the enhanced removal of both heavy metals and PAHs from low-permeability soils.

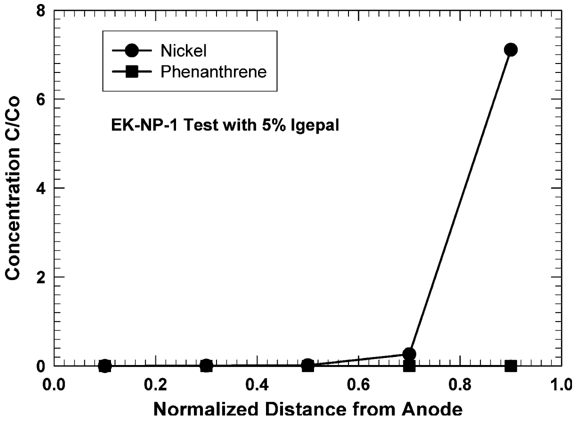


Fig. 6 Simultaneous removal of phenanthrene and nickel during electrokinetic remediation using surfactant (Igepal)

Recently, a combined electrokinetics and Fenton-like oxidation has been proposed to degrade organic contaminants within the soils and remove only heavy metals out of the soils (Reddy and Karri 2006). To assess this approach, batch and bench-scale electrokinetic experiments were conducted using kaolin soil spiked with mixed contaminants: phenanthrene as a representative PAH and nickel as a representative heavy metal. Batch experiments showed that significant oxidation of phenanthrene can be achieved using hydrogen peroxide (H_2O_2) with native iron in the soil as a catalyst. Using 5% and 30% H_2O_2, a corresponding 75% and 85% of phenanthrene was oxidized.

Electrokinetic experiments conducted using H_2O_2 at different concentrations with a 1 VDC/cm voltage gradient showed that with an increase of concentration of oxidant, an increased oxidation of phenanthrene could be achieved. Contrary to the batch experiments, using 5% and 30% H_2O_2 resulted in 27% and 56% oxidation of phenanthrene in the soil, respectively (Fig. 7a). In all electrokinetic experiments, nickel migrated towards the cathode, but it precipitated as nickel hydroxide near the cathode due to high pH conditions (Fig. 7b). The residual leachable native iron in the soil was significant indicating that native iron was not a limiting factor for the catalytic reaction of H_2O_2 to oxidize phenanthrene. Optimization of electric potential and oxidant dosage is necessary to increase phenanthrene oxidation and controlling pH near cathode is necessary to enhance removal of nickel from the soil (Reddy and Karri 2006).

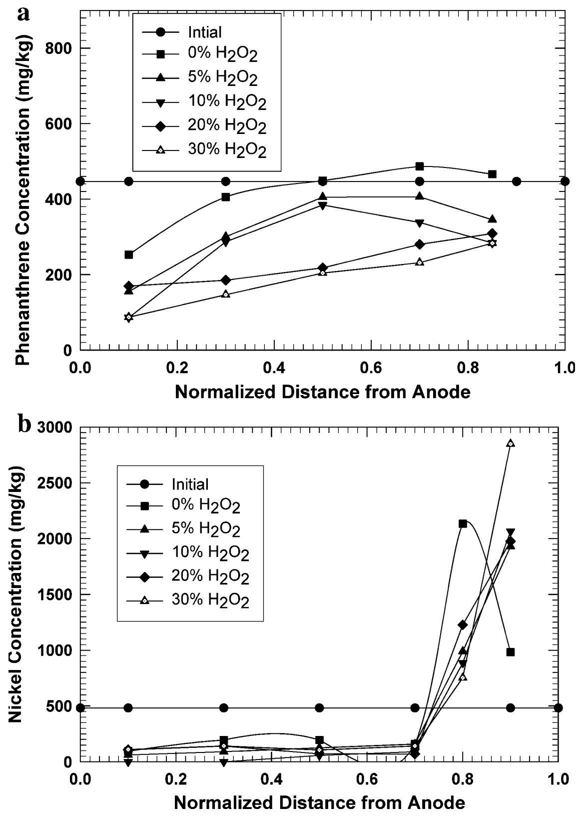


Fig. 7 (**a**) Residual phenanthrene distribution after integrated electrokinetic Fenton-like oxidation treatment. (**b**) Residual nickel distribution after integrated electrokinetic Fenton-like oxidation treatment

4 Conclusion

Although several technologies have been developed to remediate polluted sites, many of them are not applicable for sites containing low-permeability soils, heterogeneous soils, or mixed contaminants. Electrokinetic remediation technology has great potential for in-situ remediation of low-permeability and/or heterogeneous soils that have been contaminated by organics, heavy metals, or a combination of these contaminants. Electrokinetic remediation can be easily integrated with conventional remedial systems to enhance remedial efficiency and decrease the overall cost. However, electrokinetic remediation is highly dependent on site-specific geochemical conditions such as soil composition, native electrolytes, contaminant aging, and contaminant mixtures. Several research studies have been undertaken to develop fundamental geochemical characterization and enhance geochemistry for effective electrokinetic remediation. These studies have provided innovative integrated electrokinetic approaches for effective remediation of challenging polluted sites. Many of these studies were limited to bench-scale studies, and more field studies are needed to determine cost and effectiveness in field applications.

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