

# LJMU Research Online

Faisal, AAH, Rashid, IT, Hassan, WH, Al-Ansari, N, Al-Hashimi, O and Ghfar, AA

Remediation of Silty Clay Soil Contaminated with Metal lons by Electric Field Technology with the Support of Acidic Injection Wells

http://researchonline.ljmu.ac.uk/id/eprint/21891/

Article

**Citation** (please note it is advisable to refer to the publisher's version if you intend to cite from this work)

Faisal, AAH, Rashid, IT, Hassan, WH, Al-Ansari, N, Al-Hashimi, O and Ghfar, AA (2023) Remediation of Silty Clay Soil Contaminated with Metal Ions by Electric Field Technology with the Support of Acidic Injection Wells. Water, Air. & Soil Pollution. 234 (6). ISSN 0049-6979

LJMU has developed LJMU Research Online for users to access the research output of the University more effectively. Copyright © and Moral Rights for the papers on this site are retained by the individual authors and/or other copyright owners. Users may download and/or print one copy of any article(s) in LJMU Research Online to facilitate their private study or for non-commercial research. You may not engage in further distribution of the material or use it for any profit-making activities or any commercial gain.

The version presented here may differ from the published version or from the version of the record. Please see the repository URL above for details on accessing the published version and note that access may require a subscription.

For more information please contact <a href="mailto:researchonline@ljmu.ac.uk">researchonline@ljmu.ac.uk</a>

http://researchonline.ljmu.ac.uk/

# Remediation of Silty Clay Soil Contaminated with Metal Ions by Electric Field Technology with the Support of Acidic Injection Wells

Ayad A. H. Faisal · Ibtihal T. Rashid · Waqed H. Hassan · Nadhir Al-Ansari · Osamah Al-Hashimi · Ayman A. Ghfar

Abstract This work aims to propose a new approach for enhancement the reclamation of soil spiked with metal ions, specifically lead and chromium by applying electro-kinetic method. Different tests were carried out on Iraqi silty clay soil that had been polluted with either lead or chromium in concentration of 1500 mg/kg (single system) and in the form of binary system consisted of lead and chromium at concentrations of 750 mg/kg for each metal. The tests have been conducted with 7 days processing time and 1-V/cm voltage gradient. In order to improve the electro-kinetic remediation ability to remove the aforementioned metal ions from Iraqi soil, acetic acid enhanced by two injection wells were applied. Experimental outcomes proved that the lead and chromium can be removed with efficiencies of

A. A. H. Faisal (⊠) · I. T. Rashid Department of Environmental Engineering, College of Engineering, University of Baghdad, Baghdad, Iraq e-mail: ayadabedalhamzafaisal@yahoo.com; ayad.faesal@ coeng.uobaghdad.edu.iq I. T. Rashid e-mail: ibtihaltarik@yahoo.com

W. H. Hassan College of Engineering, University of Warith Al-Anbiyaa, Kerbala, Iraq e-mail: waaqidh@uokerbala.edu.iq

W. H. Hassan
Department of Civil Engineering, College of Engineering, University of Kerbala, Kerbala 56001, Iraq
N. Al-Ansari (⊠)
Department of Civil, Environmental and Natural
Resources Engineering, Lulea University of Technology, 97187 Lulea, Sweden email: nadhir.alansari@ltu.se 18.5 and 12.5%, respectively for tests performed using the distilled water. Utilizing 1-M acetic acid led to an increase in the removal efficiencies to 37 and 21.5%, respectively. However, addition of two injection wells in combination of an acetic acid result in significant increasing in the removal to be  $\leq$  59%. Finally, the removals of lead and chromium have values of 42 and 28% respectively in the binary system for acetic acid and two injection wells. These values are lower than the efficiencies for same metals in the single system operating in the same conditions, which may be due to metals competing for desorption from the soil.

**Keywords** Electric field technology · Remediation · Chromium · Acetic acid · Injection wells

O. Al-Hashimi

School of Civil Engineering and Built Environment, Liverpool John Moores University, Liverpool L3 3AF, UK e-mail: O.A.AlHASHIMI@2020.ljmu.ac.uk

A. A. Ghfar

Department of Chemistry, College of Science, King Saud University, P.O. Box 2455, Riyadh 11451, Saudi Arabia email: aghfar@ksu.edu.sa

#### **1** Introduction

The pollution of agricultural soils with hazardous metals constitutes a worldwide threat to human health and the secure cultivation of food. In agricultural ecosystems, human activities such as excessive fertilizer usage contribute to heavy metal contamination. Furthermore, acid rain or prolonged fertilization can reduce soil pH and increase soil acidity, resulting in increased bioavailability of metals in the soil and posing a risk to human health (Luan et al., 2022; Tabelin et al., 2018).

The issue of soil pollution resulted from improper disposal and handling of hazardous wastes is faced by all countries. Continuously, a wide range of financial, scientific, and technical resources are allocated for protection of environment and recovery of contamination. Technologies for remediation of contaminated soils can retain, isolate, or destroy of contaminants. Bioremediation, thermal desorption, soil vapor extraction, phytoremediation, permeable reactive barrier, or soil washing are familiar remediation techniques that applied to eliminate the chemical species from soilgroundwater system (Faisal & Ahmed 2014; Faisal et al., 2022; Faisal & Nassir, 2016; Hussain et al., 2021; Liu et al., 2018; Naji et al., 2020; Rashid & Faisal, 2018; Wang et al., 2021).

Alternative technologies have been developed as a result of the growing demand for soil remedial measures that are feasible and cost-effective; especially those used in polluted locations. Electrokinetic remediation (EKR) is the most promising emerging in situ techniques for remediated of polluted sites. EKR has the following advantages over traditional methods of remediation (Reddy, 2010): (1) simplicity with minimum requirements for equipment; (2) safety, contaminants cannot reach to the persons in the vicinity; (3) applicable for soil, sludge, sediment, and groundwater; (4) applicable for inorganic, organic, and radionuclides chemical species; (5) flexibility, can apply for insitu and ex-situ system as well as this technique can be implemented together with other techniques like bioremediation; and (6) cost-effectiveness, needs low electrical energy. Electro-kinetic phenomena that take place when electrodes are inserted into polluted soil to apply an electric field to saturated soil are the foundation of the EKR method. The

conductive medium is the pore liquid. The electrical field can enhance the movement of the liquid and; accordingly, dissolved contaminants must be transported to the electrodes. The collected chemical species in the electrode compartments are removed by separation processes like electrodeposition, chemical precipitation, and ionic membranes (Guo et al., 2020; Huang et al., 2018).

The primary components of electro-kinetic remediation involve the installation of electrodes into contaminated soils and using a low electrical field in the anodes and cathodes. Electro-migration, electrophoresis, and electro-osmosis have the potential to influence the movement of metals to the electrodes. Electro-osmosis refers to the transport of water molecules in the soil pores towards the cathodes, while electro-migration is the transfer of ionic species to electrodes with opposing charges. Electrophoresis involves the migration of particles within an electrical field and can encompass all charged molecules, such as colloids, clay grains, and organic substances (Faisal & Hussein, 2013; Reddy & Karri, 2006). During the EKR process, the water electrolysis is the predominant for transfer of electron as explained in the following equations (Faisal et al., 2022):

$$4H_2O + 4e^- \rightarrow 2H_2(g) + 4OH^- (cathode)$$
(1)

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

The gasses, like oxygen at anode and hydrogen at cathode, are emitted into the atmosphere. The hydrogen and hydroxyl ions must move to the electrodes having opposite charges. Depending on the extent of such ions along the soil, significant variations in the soil pH are occurred. Acidic conditions (low pH) are generated adjacent to the anode and alkaline conditions can occur in the region beside the cathode. The soil pH changes are influenced on the adsorption-desorption, oxidation-reduction, and precipitation-dissolution. The lower pH in anode side can cause the solubilization of cationic species enhancing their electro-migration to the cathode. However, the higher pH in cathode side can precipitate and/or adsorb of such metals, hindering electro-migration (Hosseini et al., 2011).

Several studies have shown that the use of horizontal DC in EKR can effectively remove metals from low-permeability soils (Chen et al., 2011; Ouhadi et al., 2010). An experimental study was conducted to examine the clean-up of Pb, Cu, Cd, As, and Zn, and provide information on the direction of electroosmotic flow, soil pH, and the effects of citric acid (Lee et al., 2003). The electroosmotic flow, contamination area, and cleanup efficiency are significantly impacted by metal polarity, soil type, and processing duration (Al-Hamdan & Reddy, 2008). The effects of various voltage gradients, enhancement solutions, and soil types were also investigated (Cameselle & Pena, 2016; López Vizcaíno et al., 2018; Zhang et al., 2017). An experimental investigation was performed to identify the efficacy of EKR method in the cleanup of copper from different kinds of Iraqi soil (Faisal & Hussein, 2015). The influences of the purging solution, washing process, and inclination angle of topography on the efficiency of EKR method were studied to treat of soil contained Ni, Zn, and Pb (Faisal et al., 2022). This study is to investigate the efficacy of lead and/or chromium decontamination from Iraqi lowpermeable contaminated soil by electro-kinetic process supported with unique injection wells to enhance the contaminant mobility.

# 2 Materials and Methods

#### 2.1 Soil

Naturally porous soil from Iraq was utilized. The soil was meticulously cleaned, dried, and sorted, with further sieving to ensure uniformity. Preparation and characterization of the soil are essential to guarantee high precision in the conducting of practical experiments. According to ASTM D 2974, the soil had an organic content of 0.28%, an electrical conductivity (EC) of 4500  $\mu$ S/cm, a C aCO<sub>3</sub> content of 25.8%, a pH of 8.5, a porosity of 0.49, and a capacity of cation exchange of 21.4 meq/100 g. According to ASTM D 422, the distribution analysis of particle size indicated that the soil consisted of 49% clay, 49% silt, and 2% sand; therefore, it is referred to as "silty clay."

#### 2.2 Contaminants

Pb(NO<sub>3</sub>)<sub>2</sub> and Cr(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O solutions, which were produced by BHD limited Poole in England, were used to simulate the contamination of the soil with lead and chromium. To get a representative concentration, the prepared solutions were added to the specimen. The atomic weight of lead ions is 207.20 g/ mole, while the molecular weight of Pb(NO<sub>3</sub>)<sub>2</sub> is 331.21 g/mole. For instance, the 2.397 g of Pb(NO<sub>3</sub>)<sub>2</sub> must be mixed with 1 kg of dry soil in 0.5 L of distilled water to obtain a soil sample with a water content of 50% and containing 1500 mg/kg of lead. Also, calculating the necessary mass of Cr(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O to prepare the artificially contaminated soil is depended on its molecular weight which equals 400.15 g/mole.

# 2.3 Purging Solutions

The performance of EKR process in the treatment of polluted soil must be evaluated using the familiar purging solution (PS) which represented by distilled water (DW). Experiments conducted with DW form the baseline (or reference) for other experiments implemented with an enhancement solution. This solution is used to control soil chemistry, promote solubilization, and transport contaminants in alkaline soils. The enhancement solution, similar to acetic acid (AA), is used in the cathode compartment. Acetic acid, with the chemical formula C H<sub>3</sub>COOH, is a monoprotic acid. In an aqueous solution, it partially dissociates into acetate ions (CH<sub>3</sub>COO-) and (H<sup>+</sup>) ions. While acetate ions may form complexes with metal ions in the solution, H<sup>+</sup> can lower the pH of solution and dissolve the precipitates. The use of AA to depolarize hydroxide ions produced by cathodic electrolytic reduction has several advantages including the following: (1) most metal acetates are highly soluble; (2) due to AA's high pKa value, the ion concentration generated by acid dissociation is low, resulting in a minimal increase in soil electrical conductivity; (3) it is biodegradable and environmentally safe; and (4) acetate ions also prohibit the generation of additional insoluble salts near the cathode, avoiding the formation of a low electrical conductivity zone and reducing the dissipation in the electrical energy (Reddy & Chinthamreddy, 2003; Yeung, 2006).

#### **3** Electro-Kinetic Tests

# 3.1 Electro-Kinetic Setup

The schematic representation of the EK setup (Fig. 1) is composed of cell (30-cm length, 5-cm width, and 10-cm depth), two electrode compartments, a

multi-meter. The electrodes received a constant voltage from a DC power source. To promote the propagation of the acidic front, two perforated tubes with an outer and inner diameters of 2.5 and 2 cm, respectively were inserted vertically into the soil sample until they reached the base of the cell, as shown in Fig. 1.



Fig. 1 Schematic arrangement of electro-kinetic setup used in the present study

multi-meter, and DC power supply (LODESTAR LP3005D). The actual length (L) of the soil sample inserted in the cell is 20 cm. The EK cell is exposed to the atmospheric ambient to release of gasses generated through the water electrolysis. The electrode compartment is included graphite electrode, filter paper, and valve for controlling the flow of PS.

Two graphite electrodes with cylindrical shape (chemically unresponsive to electrolysis reactions) are applied to represent the anode and cathode. Graphite is inert material which can prevent the generation of additional chemicals that may complicate the treatment process. The usage of graphite for the anode electrode is very important to resist the corrosive conditions resulted from the oxidation reaction that occurs within the anodic compartment (Acar & Alshawabkeh, 1993). The electrodes have a diameter of 2.5 cm and a length of 10 cm. During the test, the current and voltage through the soil specimen were measured with a 3.2 Testing Approach

each electro-kinetic experiment, the soil In specimens were artificially contaminated with lead or chromium. The desired concentration of 1500 mg/ kg was achieved by dissolving weighed quantities of Pb(NO<sub>3</sub>)<sub>2</sub> and/or Cr(NO<sub>3</sub>)<sub>3</sub>.9H<sub>2</sub>O in DW. A steel spatula was then used to thoroughly mix the metal-adopted water with 1 kg of dry soil in a polyethylene container. To simulate typical field moisture conditions, 500 mL of DW with a moisture content of 50% was added. The contaminated soil was then compacted uniformly with a wooden rod and placed in layers in the EK cell. The exact mass of the tested soil must be determined and situated in the cell of electro-kinetic setup. Thereafter, this soil must allow to equilibrate for 24 h to ensure the occurrence of uniform redistribution for target metal ions (Faisal & Hussein, 2015). To keep solid particles from escaping into the electrode compartments, filter

papers of the type Whatman No. 1 must be inserted at the soil sample's ends. To prevent the generation of a hydraulic gradient through the sample, the electrode compartments were filled to the same level with a pH-stabilized solution (Faisal & Hussein, 2015). The cell and power supply were then connected to apply 1-V/cm voltage gradient. Throughout the experiment, the pH, water flow, and electric current at electrode compartments were monitored for different time steps. The test was concluded when the PS flow did not change significantly or the current stabilized.

Beyond 7 days of processing, samples of solution from electrode compartments must be withdrawn. Then, the specimen of soil must be divided into 5 sections and each section was weighed and keeping in a glass container. Ten gram of soil from each section was mixed with 30 mL of DW in a glass vial and thoroughly agitated by the hand for several minutes. Then, the mixture was left for 1 h to settle the solid particles (Hansen et al., 2007). The pH of the soil samples (in terms of supernatant) and water from the electrode compartments must to measure by pH meter (WTW pH 315i, Germany). To ensure that the test results were accurate, the following precautions must be adopted: (1) using new filter papers and electrodes each test; (2) washing the EK setup with diluted acid solution for 24 h and then by DW to prevent crosspollution between the tests.

#### 3.3 M easurement of Metal Concentration in Soil

The extraction of metal ions from various sections of soil was conducted through acid digestion following the Haswell's method (Haswell, 1991). This method was applied to determine the total concentration of the adopted metals. A crushed soil sample (1 g from each section of soil) must mix with 15 and 5 mL of concentrated HCl and H NO3 respectively in a 250mL beaker. Sandy bath was used for heating the resulted mixture for duration not exceeding 60 min until the vanishing of brown vapors. Afterwards, 5 mL of concentrated HCl must add to the beaker and left at laboratory temperature for 5 to 10 min. The beaker was then warmed in the same bath once more and cooled, and 5 mL of concentrated HCl and 50 mL of heated DW were utilized to clean the beaker sides of the remaining

sample. The mixture was brought to its boiling point for 2 to 3 min. The solids were separated from the liquid using the Whatman filter paper No. 42. The precipitates were then washed with DW, and the resulting water was stored in the flask to bring the volume up to 100 mL. The metal concentration can be determined by an atomic absorption spectrophotometer (AAS) type (GBC 933 plus, Australia).

# 4 Results and Discussion

Plan of EKR experiments that applied in the present investigation for operation time of 7 days includes 7 tests. The tests EK-1, EK-2, and EK-3 are aimed examine the lead removal (with initial to concentration of 1500 mg/kg) from contaminated soil under unenhanced condition using DW, enhancement by AA, and enhancement by AA plus two injection wells, respectively. The DW and 1 M AA ( $\sim$  3) in the anodic and cathodic compartments have been utilized. For soil contaminated with Cr ions, tests designated EK-4, EK-5, and EK-6 were conducted under same previous conditions. The last test (EK-7) was implemented to investigate the treatment of soil contaminated together with Pb + Cr ions under acetic acid and two injection wells enhancement.

#### 4.1 Unenhanced Condition

For EK-1 and EK-4 tests, the PS used in the electrode compartments was DW which can represent the unenhanced condition. Results signified that the current was increased from 34 mA at the beginning of the test to reach 60 mA (EK-1) and 90 mA (EK-4) at about 24 h beyond starting of the tests with identical maximum current densities of 1.52 and 2.29 mA/  $cm^2$ . Then, these values were decreased to stabilize at value not less than 16 mA. Also, the results signified that the volumes of electro-osmotic flow collected during the EK-4 test were increased with time to be 24.5 mL beyond 48 h; however, these values remained constant until the end of this test as plotted in Fig. 2a. This figure certifies that the average electro-osmotic velocities observed for Cr (EK-4) test was  $8.92 \times 10^{-6}$  cm/s. The volume of electro-osmotic flow and average

velocities of electro-osmotic in the EK-1 have the same variation of EK-4 with maximum volume and average velocity of 50 mL and  $3.7 \times 10^{-5}$  cm/s, respectively. The range of velocity values obtained in the present experiments is following the previous findings (Reddy & Chinthamreddy, 2003). The low magnitudes of electro-osmotic flow indicate that the transport of metals is primarily governed by the electro-migration mechanism, with only a minimal role played by electro-osmosis (Kim et al., 2002).

Measurements proved that the pH values in the anode compartment for unenhanced tests were decreased from 6.8 to be 2 due to the formation of  $H^+$  ions; in contrast, the generation of  $OH^-$  ions led

**Fig. 2** Variations of electroosmotic flow (**a**) and pH plus and Cr ions distribution (**b**) for electro-kinetic of soil under un- enhancement conditions

to an increase in pH to approximately 12.5 in the cathode compartment. The concentrations of metal ions (Pb and Cr) with variation of soil pH along the soil specimen beyond the 7 days were measured. Figure 2b demonstrates that the initial Cr concentration (1500 mg/kg) in the EK-4 was decreased in the sections of 0.1, 0.3, 0.5, 0.7 (L) and accumulated at section of 0.9 (L). It is clear that the remaining concentrations of Cr were = 1255 mg/kg near the anode and = 1600 mg/kg nearest the cathode after 7 days with removal total removal efficiency not greater than 12.5%. For EK-1, the remaining concentrations of Pb were equal to 1110,



1120, 1294, 1245, and 1350 for aforementioned sections to obtain removal efficiency of 18%.

The metal concentrations nearest the anode electrode are below the initial concentration, indicating that metal ions have been transported to accumulate beside the cathode electrode. This behavior is influenced directly by the trend of soil's pH. The propagation of acidic front from anode side is faster than the migration of alkalinity plume from cathode compartment in the soil sample. This is because the mobility of hydrogen ions (=  $13.05 \times$  $10^{-4}$  m<sup>2</sup>/V h) forms approximately 1.76 times of hydroxyl mobility (=  $7.41 \times 10^{-4} \text{ m}^2/\text{V}$  h) and; consequently, this can enhance the solubilization of metal ions in the sections nearest the anode side (Faisal & Hussein, 2015). Precipitation of chemical species in the soil parts nearest the cathode electrode (alkaline conditions) can be hindered the migration of metal ions and an accumulation for such metals can be occurred. This is the main cause for low removal efficiency of metal ions from soil body (Reddy & Chinthamreddy, 1999).

# 4.2 E nhancement with Acetic Acid

The precipitation of metals within the soil nearby the cathode significantly affects the cleanup of such metals when using DW in both electrode compartments as explained in previous section. Hence, 1 M AA can use as PS in the cathodic compartment to overcome this fixation mechanism. The hydrogen ions produced from AA can play a remarkable role in the lowering of solution pH and dissolving the metals precipitates. The quantity of AA had determined to keep the cathode compartment's pH in the range 2.5–3.

For EK-5 experiment, the influence of AA presence in the cathode compartment on the variations of current density, current, electro-osmotic velocity, and electro-osmotic volume through the soil specimen contaminated with Cr are depicted in Figs. 3a and b. The current can reach a higher value of 140 mA after 30 h and then it is dropped to stabilize at 72 mA beyond 144 h. The same trend also recognized for current in the EK-2 for soil spiked with lead ions with maximum value of 89 mA occurred at 96 h. However, the current density values for two tests were smaller than 4 mA/cm<sup>2</sup>. The greatest volume of flow collected

from soil specimen contaminated with Cr is equal to 116 mL beyond 7 days with trend illustrated in Fig. 3b. This figure proves that the electro-osmotic velocities can raise rapidly to  $6.66 \times 10^{-5}$  cm/s within the first 3 h; thereafter, the velocities reduce versus the time until to became  $7.67 \times 10^{-6}$  cm/s with mean velocity of  $2.44 \times 10^{-5}$  cm/s. The same trend for variations of electro-osmotic volume and velocity was observed in EK-2 where collected volume and maximum electro-osmotic velocity having values of 125 mL and  $7.04 \times 10^{-5}$  cm/s.

The soil pH and distributions of chromium ions along the specimen at the end of the treatment period (i.e., 7 days) are depicted in Fig. 3c. Even though the cathode compartment maintains a pH close to 3, the soil's pH reached 8 nearest the cathode. The cathodic reactions may not completely depolarize through processing duration as indicated by these pH values. In contrast to the unenhanced condition, the usage of AA in the cathode compartment resulted in remarkably higher metal removal and its migration toward the cathode. Because of the lowering of pH nearest the anode, the chromium introduced into the soil will be as C  $r^{3+}$  after the voltage gradient application; thus, the Cr<sup>3+</sup> migrates toward the cathode. However, the introduction of AA can increase the velocity of electro-osmotic, increasing Cr removal efficiency to be 21.5%. Also, the measurements proved that the residual Pb concentrations along 0.1, 0.3, 0.5, 0.7, and 0.9 (L) were 609, 1010, 1127, 1183, and 758 mg/kg respectively with total removal efficiency of 37%.

4.3 Enhancement with Acetic Acid and Injection Wells

Two acidic injection wells in combination with application of acetic acid in cathode compartment were proposed and implemented in this study to improve the removals of metals through increasing their solubilization. The changes in the electrical current and electro-osmotic flow over time are illustrated in Fig. 4a and b for EK-6 (soil contaminated with Cr). The injection of acetic acid during this test may have led to the initial high current of 68 mA. Then, the current was increased to reach 150 mA beyond 24 h; then, it can slightly increase to stabilize around 160 mA at the end of the test. The flow of the electroosmotic process is directly proportional to the ionic species present in the pore liquid and; consequently, to the current magnitude, which explains why the total amount of water collected at the end of the test (beyond 7 days) reached 247 mL. The same trend was recorded for current and electro-osmotic flow in the soil contaminated with lead ions (EK-3). The highest values of current and flow volume can be occurred at the end of test to have values of 160 mA and 312 mL, respectively. This significant difference in the collected water compared to the unenhanced tests can be attributed to the use of injection wells to replenish the soil-liquid regime with PS and reduce the soil's pH. The lower pH enhances the solubilization of metals/ salts, increasing the ionic concentration, current, and water volume of electro-osmosis.

The concentrations profiles of metal ions in the soil at the end of tests EK-3 and EK-6 are plotted in Fig. 4c. The results indicate a reduction in the concentration of contaminants compared to the initial concentration. The effects of soil pH changes on the migration of these metals are also shown in the same figure. The injection wells in the EK-3 and EK-6 were found to be effective in driving the acidic front toward

Fig. 3 Variations of current

(a), electroosmotic flow (b), and pH with Cr ions distribution (c) for electro- kinetic of soil under enhancement with acetic acid



Fig. 4 Variations of current (a), electroosmotic flow (b), and pH with metal ions distribution (c) for electro-kinetic of soil under enhancement with acetic acid and injection wells

![](_page_10_Figure_1.jpeg)

![](_page_11_Figure_0.jpeg)

the negative electrode, which reduced the soil pH and increased the migration of contaminants. This figure demonstrates that the acidic plume was able to run the full length of the tested soil due to the enhancement provided by the injection wells. This decrease in soil buffering capacity resulted in a higher amount of contaminants being removed from the soil specimen, as well as a decrease in the presence of carbonate and calcite which accumulated on the surface of cathode electrode as explained by Fig. 5. The formation of an acidic front can therefore contribute to a relatively high level of contaminant removal.

#### Normalized distance from anode (x/L)

4.4 Removal of Metals and Electro-Osmotic Permeability

For each of the previous tests, the residual metal masses in the soil have been utilized to find the removal efficiencies. For EK-1 to EK-3, the lead removals were 18, 37, and 59%, respectively. However, the chromium is removed with percentages of 12.5% in EK-4, 21.5% in EK-5, and 42% in EK-6. When using 1 M AA as PS

and two wells of injection within the soil body, as in the EK-3 and EK-6 tests, the metal removals for silty clay soil were improved. Using 1 M AA as PS, more metal ions were probably removed due to the soil's higher flow. Results demonstrated that the efficiency of lead removal was greater than that of chromium removal. This could be because the soil has a high capacity for the adsorption of chromium, or PS prefers, thermodynamically, to form a complex with Pb ions.

In achieved tests, the flow direction in electroosmotic is as expected from positive to negative electrodes. The accumulative volume of

![](_page_12_Picture_2.jpeg)

Fig. 5 Accumulation of the salts on the surface of the cathode electrode at the end of the EK-6 test

effluent (cathode side) at the end of these tests is explained in Table 1. Figure 4b in combination with this table signifies that the acidic wells have positive influence on the EK operation through increasing the cumulative quantity of effluents in comparison with tests free of such wells. The rates of flow can calculate from the slopes of the relations between electroosmotic flow volume with time which ranged from 0.1458 to 1.8571 mL/h for present tests (Table 1). The soil particles' zeta potential may be related to the increased flow rate. For soil specimens with a crosssectional area A and length L under

$$Q_e = \frac{\zeta \epsilon q}{4\mu} \frac{V}{L} A \tag{3}$$

Experiment (me	etal) Cumulative volume (mL	Cumulative $Q_e$ (mL/h) volume (mL)		$k_e$ (cm <sup>2</sup> /V.s)	
EK-1 (Pb)	50	0.2976	3.307 10 <sup>-6</sup>	×	
EK-2 (Pb)	125	0.7440	$8.267 \\ 10^{-6}$	×	
EK-3 (Pb)	312	1.8571	$2.063 \\ 10^{-5}$	×	
EK-4 (Cr)	24.5	0.1458	$1.620 \\ 10^{-6}$	×	
EK-5 (Cr)	116	0.6905	$7.672 \\ 10^{-6}$	×	
EK-6 (Cr)	247	1.4702	1.634 10 <sup>-5</sup>	×	

a voltage gradient V, Eq. 3 can apply to calculate the flowrate ( $Q_e$ , mL/h) resulting from the electroosmotic mechanism as follows (Kim et al., 2002):

 Table 1
 Electro-osmotic characteristics of experiments conducted with lead and chromium contaminant

Fig. 6 Variations of current, electroosmotic flow, pH and metal ions distribution for electro-kinetic of soil under enhanced with acetic acid and injection wells for (Pb + Cr) binary system

![](_page_13_Figure_1.jpeg)

![](_page_14_Figure_0.jpeg)

Weng and Huang (2004), Eq. 3 is formulated in the shape of Darcy's law:

 $Q_e = k_e i_e A$  (4) where the electro-osmotic permeability and voltage gradient are referred to as  $k_e$ and  $i_e$ , respectively. According to Eq. 4, the average  $k_e$ has values in the range from  $1.62 \times 10^{-6}$  to  $1.634 \times 1$  $0^{-5}$  cm<sup>2</sup>/V.s with voltage gradient of 1 V/cm and A of 25 cm<sup>2</sup>. Higher current means that the resistance of electro-kinetic system is relatively low resistance and this can result in a higher  $k_e$  value. The outcomes of  $k_e$ are compatible with the findings of existing researches like Weng et al. (2001). In general, the present findings certify that the enhancement by injection wells is successful in removing metals from the soil.

# 4.5 Enhancement with Acetic Acid and Injection Wells for Binary System

The current and cumulative volume of electroosmotic movement for the EK-7 test is depicted in Fig. 6 over 7 days of treatment. The soil conductivity is related to the concentration of ionic elements in the liquid within the soil pore, which is directly related to the passed current. When the ionic concentration is high, the soil's current flow will be faster. The reduction of soil pH near the anode, caused by the electro-migration of H<sup>+</sup> released at the anode, can result in an initial raise in the current. The soil conductivity was higher as more ionic contaminants were present in the pore liquid during the acidic state. As the concentration of dissolved ionic species decreased, the soil's conductivity and current also decreased. Compared to the EK-3 and EK-6 tests, the flow rate in EK-7 was significantly lower at later stages but had higher values in the initial stages. It can be seen that most of the flow occurs within 5 days of the test. The nonuniform electro-osmotic flow is caused by the varying surface charge of the soil, characteristics of the pore liquid, and potential gradient (Al-Hamdan & Reddy, 2008). The distributions of Pb and Cr ions as well as the soil's pH profile following EK-7 are depicted in Fig. 6. In the binary experiment (EK-7) for Pb and Cr, the removal efficiencies were 42% and 28%, respectively, which was lower than the removal efficiencies for a single compound system implemented with same circumstances of injection wells and PS for (K-3 and EK-6). This may be because these metals compete with one another for desorption from soil particles.

## **5** Conclusions

The results of the electro-kinetic remediation of silty clay soil contaminated with Pb and Cr using DW as PS showed low removal efficiencies of 18 and 12.5%, respectively. The precipitation of metal ions near the cathode compartment due to the rise in soil pH from the electrolysis of the PS caused a blockage in the passage of ions and resulted in a significant decrease in remediation efficacy. To ensure the movement of metal ions from the soil to the compartments, controlling the soil pH is a suitable solution. Acetic acid was used as PS in the cathode compartment to stabilize the pH at approximately 3, which resulted in an increase in removal percentages to 37% for Pb and 21.5% for Cr. The solubilization of metal ions within the soil was also increased, leading to a significant increase in electro-osmotic flow. To improve the removal efficiency, the present investigation introduces a novel approach to enhance metal solubilization and transport through the use of acetic acid and "injection wells" along the length of the soil body. This protocol positively affects the propagation of the acidic front, enabling the transport of more ions out of the soil. With this protocol, removal percentages were dramatically increased to 59% for Pb and 42% for Cr, accompanied by a significant increase in electro-osmotic permeability coefficient and collected volume of electro-osmotic. The removal efficiencies for the Pb and Cr metals in the binary system were less than their efficiencies in the single system, which may be due to competition between these metals for desorption from the soil in the first system. The primary recommendation of this study is to conduct an optimization study for the EKR process that is related to the concentrations of the purging solutions, the costs associated with their use, number of injection wells and the amount of energy consumed.

Acknowledgements We would like to gratefully acknowledge the technical support of Environmental Engineering Department/University of Baghdad provided during the present work. One of the authors (Ayman A. Ghfar) is grateful to the Researchers Supporting Project number (RSP2023R407), King Saud University, Riyadh, Saudi Arabia for the financial support. **Data Availability** The data presented in this study are included in the article. Further inquiries can be directed to the corresponding authors. **Declarations** 

**Competing Interests** The authors declare no competing interests.

#### References

- Acar, Y. B., & Alshawabkeh, A. N. (1993). Principles of electrokinetic remediation. *Environmental Science & Technology*, 27(13), 2638–2647.
- Al-Hamdan, A. Z., & Reddy, K. R. (2008). Transient behavior of heavy metals in soils during electrokinetic remediation. *Chemosphere*, 71(5), 860–871.
- Cameselle, C., & Pena, A. (2016). Enhanced electromigration and electro-osmosis for the remediation of an agricultural soil contaminated with multiple heavy metals. *Process Safety and Environmental Protection*, 104, 209–217.
- Chen, J.-L., Yang, S.-F., Wu, C.-C., & Ton, S. (2011). Effect of ammonia as a complexing agent on electrokinetic remediation of copper-contaminated soil. *Separation and Purification Technology*, 79(2), 157–163.
- Faisal, A., & Ahmed, M. (2014). Remediation of groundwater contaminated with copper ions by waste foundry sand permeable barrier. *Journal of Engineering*, 20(9), 62–77.
- Faisal, A. A. H., & Hussein, A. A. (2013). Modeling and simulation of copper removal from the contaminated soil by a combination of adsorption and electro-kinetic remediation. *Journal of Engineering*, 19(6), 695–716.
- Faisal, A. A. H., & Hussein, A. A. (2015). An acidic injection well technique for enhancement of the removal of copper from contaminated soil by electrokinetic remediation process. *Separation Science and Technology*, 50(16), 2578–2586.
- Faisal, A. A. H., & Nassir, Z. S. (2016). Modeling the removal of cadmium ions from aqueous solutions onto olive pips using neural network technique. *Al-Khwarizmi Engineering Journal*, 12(3), 1–9.
- Faisal, A. A. H., Alkhateeb, R. W., Al-Hashimi, O. A., Shubbar, A., & Saleh, B. (2022). Removal of zinc, lead and nickel from contaminated soil by electro-kinetic technology under the influence of washing and buoyance forces with different purging solutions. *Separation Science and Technology*, 57(8), 1290–1303.
- Guo, F., Ding, C., Zhou, Z., Han, F., Tang, R., Huang, G., & Wang, X. (2020). Assessment of the immobilization effectiveness of several amendments on a cadmiumcontaminated soil using Eisenia fetida. *Ecotoxicology and Environmental Safety*, 189, 109948.
- Hansen, H. K., Rojo, A., & Ottosen, L. M. (2007). Electrokinetic remediation of copper mine tailings. *Electrochimica Acta*, 52(10), 3355–3359.
- Haswell, S. J. (1991). Atomic absorption spectrometer; theory, design and applications. Amsterdam: Elsevier.

- Hosseini, M. M., Farahbakhsh, M., & Savaghebi, G. (2011). Chelate agents enhanced electrokinetic remediation for removal of lead and zinc from a calcareous soil. *International Conference on Environment Science and Engineering*, 2011, 208–211.
- Huang, T., Liu, L., Zhou, L., & Zhang, S. (2018). Electrokinetic removal of chromium from chromite ore-processing residue using graphite particle-supported nanoscale zerovalent iron as the three-dimensional electrode. *Chemical Engineering Journal*, 350, 1022–1034.
- Hussain, B., Ashraf, M. N., Shafeeq-ur-Rahman, Abbas, A., Li, J., & Farooq, M. (2021). Cadmium stress in paddy fields: Effects of soil conditions and remediation strategies. *Science of The Total Environment*, 754, 142188.
- Kim, S.-O., Kim, K.-W., & Stüben, D. (2002). Evaluation of electrokinetic removal of heavy metals from tailing soils. *Journal of Environmental Engineering*, 128(8), 705–715.
- Lee, H. S., Lee, K., Kim, S. S., & Ko, S. H. (2003). Effects of buffering capacity and citric acid in electrolyte on electrokinetic remediation of mine tailing soils. *Journal of Industrial and Engineering Chemistry*, 9, 360–365.
- Liu, L., Li, W., Song, W., & Guo, M. (2018). Remediation techniques for heavy metal-contaminated soils: Principles and applicability. *Science of the Total Environment*, 633, 206–219.
- LópezVizcaíno, R., Yustres, A., Asensio, L., Saez, C., Cañizares, P., Rodrigo, M. A., & Navarro, V. (2018). Enhanced electrokinetic remediation of polluted soils by anolyte pH conditioning. *Chemosphere*, 199, 477–485.
- Luan, Y., Xu, J., Zhou, J., Wang, H., Han, F., Wang, K., & Lv, Y. (2022). Migration and removal of labile cadmium contaminants in paddy soils by electrokinetic remediation without changing soil pH. *International Journal of Environmental Research and Public Health*, 19(7), 3812.
- Naji, L. A., Faisal, A. A. H., Rashid, H. M., Naushad, M., & Ahamad, T. (2020). Environmental remediation of synthetic leachate produced from sanitary landfills using low-cost composite sorbent. *Environmental Technology & Innovation*, 18, 100680.
- Ouhadi, V. R., Yong, R. N., Shariatmadari, N., Saeidijam, S., Goodarzi, A. R., & Safari-Zanjani, M. (2010). Impact of carbonate on the efficiency of heavy metal removal from kaolinite soil by the electrokinetic soil remediation method. *Journal of Hazardous Materials, 173*(1–3), 87–94.
- Rashid, M. H., & Faisal, A. A. H. (2018). Removal of dissolved cadmium ions from contaminated wastewater using raw scrap zero-valent iron and zero valent aluminum as locally available and inexpensive sorbent wastes. *Iraqi Journal of Chemical and Petroleum Engineering*, 19(4), 39–45.
- Reddy, K. R. (2010). Technical challenges to in-situ remediation of polluted sites. *Geotechnical and Geological Engineering*, 28(3), 211–221.
- Reddy, K. R., & Chinthamreddy, S. (1999). Electrokinetic remediation of heavy metal-contaminated soils under reducing environments. *Waste Management*, 19(4), 269– 282.
- Reddy, K. R., & Chinthamreddy, S. (2003). Sequentially enhanced electrokinetic remediation of heavy metals in low

buffering clayey soils. Journal of Geotechnical and Geoenvironmental Engineering, 129(3), 263–277.

- Reddy, K. R., & Karri, M. R. (2006). Effect of voltage gradient on integrated electrochemical remediation of contaminant mixtures. *Land Contamination & Reclamation*, 14(3), 685– 698.
- Tabelin, C. B., Igarashi, T., Villacorte-Tabelin, M., Park, I., Opiso, E. M., Ito, M., & Hiroyoshi, N. (2018). Arsenic, selenium, boron, lead, cadmium, copper, and zinc in naturally contaminated rocks: A review of their sources, modes of enrichment, mechanisms of release, and mitigation strategies. *Science of the Total Environment*, 645, 1522–1553.
- Wang, Y., Li, A., & Cui, C. (2021). Remediation of heavy metalcontaminated soils by electrokinetic technology: Mechanisms and applicability. *Chemosphere*, 265, 129071.
- Weng, C.-H., & Huang, C. P. (2004). Preliminary study on treatment of soil enriched in chromite ore processing residue by electrokinetics. *Practice Periodical of Hazardous, Toxic, and Radioactive Waste Management*, 8(2), 67–72.
- Weng, C. H., Huang, C. P., & Sanders, P. F. (2001). Effect of pH on Cr(VI) leaching from soil enriched in chromite ore processing residue. *Environmental Geochemistry and Health*, 23(3), 207–211.
- Yeung, A. T. (2006). Contaminant extractability by electrokinetics. *Environmental Engineering Science*, 23(1), 202–224.
- Zhang, Y., Huang, T., Huang, X., Faheem, M., Yu, L., Jiao, B., Yin, G., Shiau, Y., & Li, D. (2017). Study on electrokinetic remediation of heavy metals in municipal solid waste incineration fly ash with a three-dimensional electrode. *RSC Advances*, 7(45), 27846–27852.