The Combined Electrokinetic and Flushing Remediation of Multiple Heavy Metals Co-Contaminated Soil Enhanced with Acid Treatment

Jun Hou,1 Li Fan,1* Mingyuan Zhou,12 Xuefeng Zhu,12 Xiuli Li,12 Jie Guan,12 Guilan Gao12 and Fengying Fu1

Electrokinetic remediation (EK) is widely regarded as a green technology applied to deal with the contaminated soil. However, the treatment efficiency of EK is particularly dependent on the desorption of contaminants from the soil. In this work, the electrokinetic remediation of soil co-contaminated with Cd, Cu and Pb enhanced with acid pre-treatment and the EDTA-2Na flushing was studied. A series of experiments were conducted to investigate the removal behavior of heavy metals from soil with different voltage gradients, electrode materials and concentrations of EDTA-2Na. The results included the following main aspects: 1) Under the voltage gradient of 1.0 V/cm, the removal efficiency of Cd, Cu and Pb reached 91.09 %, 80.34 % and 72.38 %, respectively. Compared to the other studies, the electric current was particularly high since the soil was pre-acidified. 2) The residual form of heavy metals in the soil was transformed to the other fractions easier to be removed due to the function of EK and EDTA-2Na flushing. 3) The removal of Cd, Cu and Pb was mainly influenced by the electromigration under this experimental condition. A combined electrokinetic and flushing remediation enhanced with acid treatment process was found to be effective for Cd, Cu and Pb removal simultaneously.

Keywords: Electrochemistry; Environmental remediation; Heavy metals; Pollution control

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1. Introduction

Soil provides human with natural resources for surviving and developing. However, the soil contamination has been a serious problem threatening the public health and ecological safety, and the soil remediation technologies have been paid more and more attention. Bioremediation, soil flushing, solidification/stabilization, phyto remediation and electrokinetics have been the most widespread methods of treating polluted soil.1

Nowadays, the co-contamination of soil by heavy metals and organics is a complex problem, hence the application of treatment process to deal with different types of contaminants existing in soil is considerable.2 Among the available treatment technologies, EK deserves particular attention due to its superiority. It has shown a potential promise in situ and ex situ treating fine and low-permeable co-contaminated soil.3 Under a fixed directed current electric field, there are two main remediation mechanisms imposed on the contaminants redissolving and migrating into water. In particular, ionic pollutants electromigrate towards the electrode of opposite charge and neutral pollutants in the pore fluid can be transported into electrolyte by electroosmosis.4 EK achieved consolidation, dewatering and removal of salts and inorganic contaminants in soil simultaneously.5 For heavy metals and organic compounds detached from soil particles, the mechanism of decontamination are mainly electromigration and electroosmotic flow (EOF) respectively. Additionally, there are other reactions during the EK process such as adsorption/desorption, precipitation/dissolution and so on.

However, EK has the limitation of remediation efficiency for the extraction and transport of hydrophobic pollutants and it just can power the migration of contaminants but unable to degrade them. Thus, lots of researchers devoted to improve and enhance the comprehensive capacity of treatment using EK and other technologies like EK-Bioremediation,6 EK-Ultrasound,7 EK-PRB8,9 and EK-Flushing10,11 to deal with various types of pollutants.

In recent works, chelating agents and surfactants have frequently been used to increase the desorption of contaminants from soil. Several studies have investigated the EK-Flushing removal of co-contaminates. Colacicco12 performed the enhanced EK remediation technology to decontaminate the heavy metal and organic compound co-contaminated soil by applying the chelating agent EDTA, with removal yields ranging from 28 % to 84 %. Alcántara13 also observed that the selection of the processing fluid and pH control were the main key variables of the EK process. They found that the addition of 1 % TW80 and 0.1M EDTA was the most effective combination treating the soil polluted with lead and phenanthrene, and under the operating conditions, more than 90% and 70% of pollutants were removed after 30 days from the kaolin clay and the sandy soil, respectively.

In this work, the electrokinetic remediation of soil contaminated with Cd, Cu and Pb combined with EDTA-2Na flushing was studied. The main objective was to achieve the simultaneous removal of multiple heavy metals from soil and study the decontamination mechanism. The effects of operating conditions such as voltage gradient, different electrode materials, and concentration of EDTA-2Na on the remediation efficiency were investigated. Furthermore, the

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removal mechanism of heavy metals was analyzed by measuring the distribution of heavy metals in soil and their quantities in the effluents from the electrode chambers.

2. Materials and Methods

2.1 Experimental soil
A natural soil was collected from the topsoil layer in campus. Soil sample was air-dried and stored for later tests. Before being used, it was ground and passed through 20-mesh sieve. The main physical and chemical characteristics of soil are summarized in Table 1.

The soil was artificially spiked with a solution of Cd(NO₃)₂·4H₂O, Cu(NO₃)₂·3H₂O and Pb(NO₃)₂ according to the target concentrations and soil moisture of 40%. The final concentrations of Cd, Cu and Pb in soil were 8.92±0.23 mg/kg, 189.56±17.53 mg/kg and 331.70±44.8 mg/kg, respectively. The soil was pre-treated by two stages of acidification, and the soil pH was 6 and the zeta potential -1.094 mV, while the electrical conductivity was 1.79 ms/cm. The soil was packed in the cell and balanced with NaNO₃ solution (0.1 M) in both sides of the electrode compartments for 24 hours.

2.2 Electrokinetic Test Setup
The schematic diagram of the electrokinetic experimental apparatus is shown in Fig. 1. The reaction system was composed of a DC power, an electric current monitoring and recording system, a reactor, a pH meter and two peristaltic pumps. The experimental reactor consisted of three compartments: the anolyte cell (80 mm×100 mm×80 mm), the soil cell (250 mm×100 mm×80 mm), and the catholyte cell (80 mm×100 mm×80 mm). The soil cell and electrode chambers were connected by flange. In order to prevent soil particles from diffusing outwards, the filter paper were placed between the soil and the electrode chambers.

2.3 Experimental Program
The parameters associated with each experiment are listed in Table 2. The operation of EK1, EK2, EK3 and EK4 were conducted with 0.01 M EDTA-2Na, while EK5 was carried out with 0.05 M EDTA-2Na to analyze the influence of different concentration of chelating agent.

Table 1 The main properties and major oxide composition of the tested soil.

<table>
<thead>
<tr>
<th>Item</th>
<th>Properties</th>
<th>Item</th>
<th>Contents(%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cd (mg/kg)</td>
<td>1.92</td>
<td>SiO₂</td>
<td>55.054</td>
</tr>
<tr>
<td>Cu (mg/kg)</td>
<td>39.56</td>
<td>Fe₂O₃</td>
<td>17.346</td>
</tr>
<tr>
<td>Pb (mg/kg)</td>
<td>51.70</td>
<td>Al₂O₃</td>
<td>13.181</td>
</tr>
<tr>
<td>pH</td>
<td>8.41</td>
<td>K₂O</td>
<td>5.795</td>
</tr>
<tr>
<td>Electrical conductivity (us/cm)</td>
<td>89.9</td>
<td>TiO₂</td>
<td>1.892</td>
</tr>
<tr>
<td>Water content (%)</td>
<td>5</td>
<td>BaO</td>
<td>0.674</td>
</tr>
<tr>
<td>Organic matter (g/kg)</td>
<td>15.5</td>
<td>MnO</td>
<td>0.333</td>
</tr>
<tr>
<td>CEC (cmol/kg)</td>
<td>26.63</td>
<td>ZrO₂</td>
<td>0.133</td>
</tr>
<tr>
<td>Zeta potential (mV)</td>
<td>-4.80</td>
<td>RbO</td>
<td>0.061</td>
</tr>
<tr>
<td></td>
<td></td>
<td>ZnO</td>
<td>0.048</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Y₂O₃</td>
<td>0.019</td>
</tr>
</tbody>
</table>

Table 2 The parameters associated with the EK experiments.

<table>
<thead>
<tr>
<th>Test</th>
<th>Anolyte</th>
<th>Catholyte</th>
<th>Voltage gradient(V/cm)</th>
<th>Anode material</th>
<th>Cathode material</th>
</tr>
</thead>
<tbody>
<tr>
<td>EK1</td>
<td>0.01M EDTA-2Na</td>
<td>0.01M EDTA-2Na</td>
<td>0.50</td>
<td>graphite</td>
<td>graphite</td>
</tr>
<tr>
<td>EK2</td>
<td>0.01M EDTA-2Na</td>
<td>0.01M EDTA-2Na</td>
<td>0.75</td>
<td>graphite</td>
<td>graphite</td>
</tr>
<tr>
<td>EK3</td>
<td>0.01M EDTA-2Na</td>
<td>0.01M EDTA-2Na</td>
<td>1.00</td>
<td>graphite</td>
<td>graphite</td>
</tr>
<tr>
<td>EK4</td>
<td>0.01M EDTA-2Na</td>
<td>0.01M EDTA-2Na</td>
<td>1.00</td>
<td>platinized titanium</td>
<td>titanium</td>
</tr>
<tr>
<td>EK5</td>
<td>0.05M EDTA-2Na</td>
<td>0.05M EDTA-2Na</td>
<td>1.00</td>
<td>platinized titanium</td>
<td>titanium</td>
</tr>
</tbody>
</table>

Fig. 1 The schematic diagram of the electrokinetic test setup.
operation of EK1, EK2 and EK3 aimed at evaluating the EK remediation of contaminated soil under different voltage gradient. The comparison of EK3 and EK4 was to investigate the applied electrode material. For all the tests, the electrolyte solutions for the anode and the cathode were fed and refreshed by two peristaltic pumps at a flow rate of 0.5 mL/min, and the catholyte pH was controlled in the range of 4 to 5 during the remediation process. The experiments were run at a constant voltage gradient for 10 days and the electric current was recorded every 10 minutes throughout the experimental period. In addition, the EOF was calculated by the difference of the volume of catholyte and anolyte overflowed every 24 h.

The soil was separated equally into five sections, and a fraction of each section was taken to determine the soil pH, the water content, the electrical conductivity and the residual Cd, Cu, and Pb concentrations after 10 days. The volumes of the effluents from the electrode cells and the concentrations of the heavy metals were analyzed daily.

2.4 Analytical methods
After the treatment, the soil samples were dried for 24 h at 50 °C, and ground with an agate mortar. Each soil sample (0.25 g) was digested with 5 mL HF and 5 mL HNO$_3$ at 100 °C for 1 h in PTFE chamber, then added with 2 mL HNO$_3$ and 2 mL HClO$_4$ at 150 °C for 1.5 h, and heated up to 220 °C until the volume is reduced to about 3 to 5 mL, and finally the liquid is diluted to a constant volume of 50 mL. After that, the samples were filtered through PESM filters (0.45 μm). The concentrations of the heavy metals were analyzed by ICP (iCAP6300, Thermo Scientific, USA). The soil pH was measured with a soil to water ratio of 1:2.5 by a pH meter (HI 99121, HANNA, China). The electrical conductivity of the soil was determined by a conductivity meter (FE30, Mettler-Toledo, China). The water content of the soil was measured by a moisture meter (HR83-P, Mettler-Toledo, Switzerland), and the electric current data by a digital multimeter (86E, Victor, China).

3. Results and discussion
3.1 Electric current
Fig. 2 shows the time course of the electric current density. The current density was proportional to the voltage gradient applied to the electrokinetic system. The effect of the voltage gradient on the removal efficiency was similar to those reported for other soils.$^{11}$ Under the voltage gradient of 1V/cm, the highest current reached 2.5 A in EK3. According to the other studies,$^{13,16}$ the electric currents were less than 1.0 A in most cases under the voltage gradient of 1V/cm. The soil acidification before the tests and pH control of the catholyte with HNO$_3$ may contribute to the high current density. The current density of EK4 was lower compared to that of EK3, since the graphite electrode used in EK3 exhibited higher electrocatalytic activity. As for EK5, the low pH of anolyte resulted in EDTA precipitation, and the electric current level was relatively low. In all experiments, even though there were obvious fluctuations and multiple peaks in the curves, the current density increased from the initial value to a maximum and then decreased gradually, similar to the up-and-down pattern reported in previous studies.$^{11}$ The high electric current values corresponded to the acidification and the refreshing of the electrolyte contributing to the movement of mobile ions. The decrease of the current density may result from the following main aspects: a) the increase of the anolyte resistance due to the dissolution of the anode of graphite plate; b) the accumulation of the impurities attached to the electrode surface in the electrode chamber.

3.2 Cumulative EOF
Fig. 3 shows the cumulative EOF during the EK treatment. In a general way, the direction of the EOF was toward the cathode based on the theory of electric double layer$^{17}$ since the zeta potential of the initial tested soil was negative.

During the EK process, the EOF may be influenced by the soil pH and the electric conductivity. Lorenz$^{2}$ reported that the zeta potential of soil was highly connected with the pH of the pore solution. The equation was listed as follows:

$$\zeta (mV) = -38.6 + 2.81 e^{-0.48pH}$$

where $\zeta$ is zeta potential and zero surface charge occurring near pH 4.0.$^{20}$ However, the mobility of protons generated at the anode was faster than that of hydroxyl ions generated at the cathode, then the acid front associated with protons propagating toward the cathode made the zero surface charge of the soil change. Moreover, the increase of zeta potential inhibited the electroosmotic velocity and reversed the direction of EOF according to the Helmholtz-Smoluchowski relation.$^{20}$

where $u_e$ is the electroosmotic velocity, $\nu$ is the permeitvity of the pore liquid, $E$, the electric field parallel to the direction of the electroosmotic flow, and $\mu$ the viscosity of the pore liquid.
3.3 Variation of soil characteristics

3.3.1 Soil moisture and temperature

The soil moisture may affect the electroremediation because it alters the electroosmotic flow rate and hence decontamination of the soil by EK process. Fig. 4 and Fig. 5 present the moisture profiles and the average temperature of the soil after the EK experiment, respectively. The moisture of most soil sections after 10 days’ reaction decreased from 40% due to the water evaporation by the heat generated during the electrokinetic remediation. Paramkusam reported that the moisture content values were low and it may be due to higher temperature gradient after the treatment. The soil temperature changed under different voltage gradient in the operation of EK1, EK2 and EK3. However, the soil moisture of these tests was not only influenced by the soil temperature but also the direction of EOF. Although the average temperature of the soil for EK3 was the highest, its soil moisture near the anode reached 40.69%. The pore solution in the soil of EK3 flowed towards the anode via EOF (Fig. 3), resulting in a higher moisture content of the soil near the anode.

\[ u_{eo} = \frac{\varepsilon \kappa E_x}{\mu} \]

The volume of the reversed EOF in EK3 increased rapidly and reached 2833 mL in the end of the test since the high electric current enhanced the movement of hydrogen ions bringing about a low pH of the soil. Similar volumes of cumulative EOF for EK1 (431 mL) and EK5 (432 mL) after 10 days of EK process were observed. The direction of the EOF was toward the cathode in EK1 while the electroosmotic velocity was retarded since the electric field strength was small. The EOF of EK5 (432 mL) was higher than that of EK4 (45 mL) owing to the different viscosity of EDTA-2Na solution used according to the Helmholtz-Smoluchowski relation. The cumulative EOF of EK2 (75 mL) increased at first and reached a peak of 836 mL, then the EOF showed a downtrend, which meant that the direction of EOF changed toward the anode.
3.3.2 Soil pH and electric conductivity

As shown in Fig. 6, the soil pH decreased compared to the initial pH value of 6. During the EK process, H\(^+\) generated at the anode migrated through the soil towards the opposite electrode and the moving rate was higher than that of OH\(^-\). Furthermore, the nitric acid added in the soil kept the soil pH at a low level, neutralizing OH\(^-\) generated from the cathode. The soil pH of EK1 ranged from 1.94 near the anode to 6.46 near the cathode, while the pH distribution of other tests fluctuated between about 1.0 and 3.0. Higher voltage gradient enhanced the movement of hydrogen ions, intensifying the soil acidification. However, the soil acidification facilitated the desorption of the heavy metals. As the soil pH decreased gradually, the zeta potential or net surface charge of soil became positive. The EOF toward the cathode decreased, and finally the direction of the flow reversed toward the anode based on the electric double layer theory.\(^7\) Thus, the desorption of heavy metals from the soil and the EOF important for the efficiency of EK treatment were both connected with the soil pH. The electrical conductivity (EC) of soil shown in Fig. 7 exhibits an increasing trend from the cathode to the anode, since the content of free ions close to the anode was higher. The curves of EC are related to the soil pH, and generally the EC of soil increases with the decrease of pH. After the EK treatment, the soil EC significantly increased compared to the initial EC of 1.79 ms/cm in all tests. The soil EC of EK3 near the anode was higher than that of the other tests due to the lowest soil pH and the dissolution of more ions in soil under higher voltage gradient.

3.4 Distribution of heavy metals in soil after electrokinetic treatment

3.4.1 The removal of Heavy Metals

The concentrations of heavy metals in the soil after EK treatments are presented in Fig. 8. With the continued conditioning of the catholyte pH by adding the nitric acid solution, the soil became acidic and most Cd, Cu, Pb remained as the cationic form. The migration of the cationic metals under the electric field showed similar behavior due to the electromigration process. The heavy metals moved toward the cathode and the concentration near the anode area was much lower than the initial value after 10 days of EK treatment. Cd\(^{2+}\) accumulated in the soil mainly at 75 mm from the cathode in EK1 under a relatively low voltage gradient of 0.5V/cm. Similarly to Cd, as the voltage gradient was elevated, the removal of Cu was enhanced and the accumulation area of Cu was closer to the cathode. The removal rate of Pb was lower than that of Cu and Cd, since Pb had strong adsorption and affinity with soil particles. According to Amrate,\(^2\) Pb was removed at a relatively low level because of strong adsorption and less dissolution. However, the removal efficiency of Pb also increased with the rise of the voltage gradient. The Pt plated titanium net was used as the anode to prevent the electrode corrosion and titanium plate was used as the cathode in EK4. However, it was observed that more heavy metals accumulated near the cathode in EK4 compared to EK3 because the cathode of titanium plate was not renewed during the experiment, and the contaminants dissolved from the cathode may re-contaminate the soil. In all experiments, EDTA-2Na was added to facilitate the desorption of heavy metals from the soil, since the formation of coordinate bond between metallic ions and EDTA-2Na is possible due to the presence of four acetate groups and two nitrogen atoms with free electron pairs. The transport of EDTA-2Na leads to the desorption of metals and the formation of new species that can be moved either by ionic migration if charged or by electroosmosis.\(^3\) The influence of the concentration of EDTA-2Na on the removal of the heavy metals was analyzed by comparing the results from EK4 and EK5. In theory, EDTA-2Na reacted with the heavy metals to form complexes increasing the removal efficiency from the soil.\(^24\)\(^\_\)\(^25\) However, it was observed that quite a few heavy metals accumulated near the cathode in EK5. There may be two main reasons: (1) the electric current of EK5 was relatively small, with the maximum of 0.6 A, resulting a lower electromigration efficiency of heavy metals and metal chelates, and (2) the EDTA-2Na precipitated due to the reduced solubility when the soil acidified\(^b\) and the precipitation of EDTA-2Na may influence the desorption of heavy metals.

In this study, Spearman correlation was applied to determine the correlation between the characteristics of soil (moisture, temperature, Table 3 Correlation coefficient (r) between removal efficiency of heavy metals and soil properties.

<table>
<thead>
<tr>
<th></th>
<th>Moisture</th>
<th>Temperature</th>
<th>pH</th>
<th>EC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Spearman Cd</td>
<td>.500</td>
<td>-.900(^*)</td>
<td>-.600</td>
<td>.800</td>
</tr>
<tr>
<td>Cu</td>
<td>.700</td>
<td>-.900(^*)</td>
<td>-.900*</td>
<td>.700</td>
</tr>
<tr>
<td>Pb</td>
<td>.200</td>
<td>-.600</td>
<td>-.500</td>
<td>.200</td>
</tr>
</tbody>
</table>

*At 95 % confidence level (P<0.05).
pH, EC) and the removal efficiency of heavy metals (Cd, Cu, Pb) for EK3. The results are presented in Table 3.

A statistically significant correlation was not detected between the soil pH and the removal efficiency of Cd and Pb, but a negative correlation at the level of $P<0.05$ was detected between soil pH and the removal efficiency of Cu ($r = -0.900$). Similar results were reported by Sungur, and in his work, linear relation was not found between soil pH and mobility of Cd and Pb, however, there was a negative correlation between the concentration of Cu and soil pH.

### 3.4.2 Distribution of the heavy metals’ speciation

Although most of the heavy metals migrated from the soil after the EK treatments, some remained in soil especially near the cathode. The speciation of heavy metals at different locations of soil was analyzed by means of sequential extraction of BCR. The total Cd, Cu and Pb in soil were sub-divided into four different fractions including residual (Resi.), oxidizable (Oxid.), reducible (Redu.), and acid extractable (Extr.) heavy metals. The pH, redox potential, and organic matter can influence the mobilisation and retention of different fractions of metals in soil.

Before the experiment, the main fraction of Cd, Cu and Pb in the soil was Resi., and it was transformed to other fractions and removed by the EK process. In the operation of EK1, EK2 and EK3, with the increase of the voltage gradient, the removal efficiency of the heavy metals except the Resi. fraction was enhanced. The Oxid. fraction accumulated in the soil near the cathode since the moisture of soil decreased. Gao reported that the accumulation of the Oxid. fraction was caused by the desiccation of soil in the EK process. The Extr. fraction of Cd accumulated near the cathode of EK1, since the soil pH was 6.46 and Cd$^2+$ may precipitated as Cd(OH)$_2$, in this area. In EK1, Cu accumulated at 75 mm from the cathode with 17.70 % Redu. fraction. The adsorption of Cu with Fe$_2$O$_3$ and MnO in soil caused the accumulation of oxide combination state. The Oxid. fraction of Cu was 25.24 % as Cu was also bound to various forms of organic matter by complexation. Most of Pb remaining in the soil after the treatment was the Resi. fraction, which was hard to remove. The Extr. heavy metals accumulated near the anode in EK5 where the soil pH was 2.69 contributing to the precipitation of EDTA-2Na that obstructed the removal of heavy metals.

### 3.4.3 Migration mechanism of heavy metals in EK process

The concentrations of Cd, Cu, and Pb in the different soil sections for EK3 are shown in Fig. 10. Most of the heavy metals migrated toward the cathode since the electromigration dominated the removal of heavy metals in the soil. Vane and Zang reported that the removal of contaminants was influenced by the electromigration and the EOF simultaneously and the ionic heavy metals were mainly affected by the electromigration. The mobility velocity of the heavy metals decreased in the order of Cd > Cu > Pb in this work. Most of Cd was removed in the first 5 days, while the migration of Cu was relatively slower. The removal efficiency of Pb was poor and part of Pb accumulated near the cathode at the end of the treatment. However, compared to other tests, the removal efficiency of Pb was relatively high in this work. The migration mechanism of heavy metals was complicated since the soil pH changed, the electroosmotic flow reversed to the anode and the anionic metal-EDTA-2Na complexes formed during the treatment. Reddy also summarized that the EDTA-enhanced EK remediation of multiple heavy metals in soils was influenced by many complicating factors, such as the chemistry and behavior of metal contaminants, the pH variation, the low mobility of EDTA, and the opposing direction of EOF.

During the EK process, the metal ions migrated toward the cathode and eventually moved into the catholyte. Fig. 11 presents the accumulated quality of Cd, Cu and Pb in the effluents from the soil during the treatment. It was obvious that the content of the heavy metals in the effluent from the cathode was significantly higher than that in the effluent from the anode, demonstrating again that the electromigration dominated the removal of heavy metals in this work. Adding the EDTA-2Na in the treatment, a variety of different complexes, such as

<table>
<thead>
<tr>
<th>Speciation (mg/kg)</th>
<th>Untreated soil</th>
<th>Soil after acidification</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Cd</td>
<td>Cu</td>
</tr>
<tr>
<td>Total</td>
<td>1.92</td>
<td>39.56</td>
</tr>
<tr>
<td>Extr.</td>
<td>0.05</td>
<td>0.35</td>
</tr>
<tr>
<td>Redu.</td>
<td>0.07</td>
<td>0.86</td>
</tr>
<tr>
<td>Oxid.</td>
<td>0.02</td>
<td>1.40</td>
</tr>
<tr>
<td>Resi.</td>
<td>1.78</td>
<td>36.96</td>
</tr>
</tbody>
</table>

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**Fig. 9** The distribution of heavy metals’ speciation from the cathode to the anode after 10 days of EK treatment.
4. Conclusion
Experiments were conducted to investigate the removal of Cd, Cu and Pb in the soil using the EK technology combining the chelating agent EDTA-2Na. From the results, EK performed a potentially effective technology for soil remediation. The main conclusions can be drawn as follows:

(1) The heavy metals were mainly removed through the electromigration and increasing the voltage gradient promoted the removal of the heavy metals effectively.

(2) Maintaining a slightly acidic soil condition using the pH control system facilitated the removal of the heavy metals.

(3) The utilization of platinized titanium mesh as the anode resisted anode corrosion, however, the electric current level was lower than that using the graphite anode resulting in relatively poorer removal efficiency.

(4) Adding EDTA-2Na enhanced the desorption of the heavy metals from the soil. However, further studies should be conducted to analyze the migration of different metal chelates during the treatment. Controlling the condition to form stable metal chelates was crucial for improving the removal efficiency of heavy metals.

(5) The Resi. fraction of metals remained in soil after the treatment due to the poor migration ability. Considering the Resi. fraction of metals was transformed to other speciations relatively easy to be removed during the EK process, the removal efficiency could be improved if given more running time.

(6) The removal efficiency of the heavy metals decreased in the order of Cd > Cu > Pb in this work. A negative correlation was found between the soil pH and the removal efficiency of Cu.

Acknowledgments
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\begin{align*}
\text{Cd}^2+, \text{CdHL}^-, \text{CdH}_2\text{L}^-, \text{CdH}_3\text{L}^2-, \text{CuL}^-, \text{CuHL}^-, \text{Cu(H}_2\text{O-L}^-, \text{Cu(OH)H}_2\text{L}^2-, \text{PbL}^-, \text{PbHL}^-; \text{(where } L = \text{EDTA}) \text{ can form depending on the solution pH.} \\
\text{So the heavy metals were detected in the anode effluent since the metal-EDTA-2Na anionic complexes migrated toward the anode and the reversed EOF may also transport the heavy metals to the anode.} \\
\text{Controlling the condition to form stable metal chelates was crucial for improving the removal efficiency of heavy metals.}
\end{align*}
\]
References