

Enhanced Electrokinetic Remediation of Heavy-Metals Contaminated Soil in presence tetrasodium N,N-bis(carboxymethyl) glutamic acid (GLDA) as chelator

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Electrokinetic remediation (EK) is an emerging method that is used to decontaminate soil contaminated with heavy metals. Heavy metal removal rates can be enhanced by applying chelators to the soil; however traditional chelators have shortcomings, such as poor biodegradability (EDTA, DTPA, etc.), high cost (EDDS, etc.), and low complexing abilities (citric acid, NTA, etc.). In this study, we used tetrasodium N,N-bis(carboxymethyl) glutamic acid (GLDA), a novel and cheap biodegradable chelator to enhance the electrokinetic remediation of a heavy metals-contaminated soil. The batch extraction experiments used three types of biodegradable chelators (lactic acid, citric acid, and GLDA) and four EK tests (T1: catholyte controlled at pH 4 without GLDA; T2: only GLDA; T3: bipolar electrolyte controlled at pH 6 with GLDA; T4: cation and anion exchange membranes with GLDA) were conducted. The batch experiments showed that the Cu and Ni extraction rates for GLDA (39% to 48%) were higher than for citric acid (26% to 41%) and lactic acid (0.44% to 25%), and the solution pH had little effect on the GLDA extraction rate. The EK experiments showed that the T3 and T4 treatments had little effect on soil pH and EC. However, T1 greatly decreased soil pH and increased soil EC. A total of 12.9%–20.1% Cu and 24.8%–27.7% Ni were removed from the soil after 8 days of the T3 and T4 treatments. Those removal rates were higher than those obtained with the T1 and T2 treatments. The total Cr removal rates in all treatments were very low (1.55% to 5.70%), which could be due to the high percentage of the residual Cr form. The results for energy consumption indicated that T4 had the lowest energy consumption (19.3 kWh t⁻¹). Therefore, installing cation and anion exchange membranes could increase Cu and Ni removal rates and decrease energy consumption. Furthermore, GLDA could potentially be used during the EK remediation of soil contaminated with heavy metals.

Keywords: Electrokinetic remediation; GLDA; Heavy metal; Ion exchange membrane; pH control

1. INTRODUCTION

Society and economic developments have meant that anthropogenic activities (including mining,

smelting, solid waste, wastewater irrigation, etc.) have led to substantial heavy metal pollution, especially in China [1-2]. In 2014, the Ministry of Environmental Protection (MEP) and the Ministry of Land and Resources of China issued a nationwide soil pollution survey communique. Of all the samples analyzed, 16.1% exceeded the environmental quality standard set by the MEP, and 82.8% were contaminated with heavy metals. In contrast to organic pollutants, heavy metals cannot be degraded by chemical or biological methods, and can migrate, transform, and accumulate in the environment, which means that they pose considerable environmental risks [2-3].

At present, the remediation technologies for soil contaminated with heavy metals mainly include landfill, solidification/immobilization [4-5], leaching [6-7], and phytoremediation [8], etc., but these technologies have certain limitations when used to remediate clayey and low-permeability soil [9-10]. Electrokinetic (EK) remediation is an in-situ remediation technique that is often used to remediate clayey and low-permeability soils [11-12]. In order to improve the efficiencies of heavy metal removal from soil under EK, a variety of enhanced remediation technologies have been applied, such as catholyte acid control [13-14], ionic membrane technology [15-16], chelate-enhanced remediation [17-18], electrode exchange, and the approaching anode technique [19]. The aim of these technologies is to improve the mobility of heavy metals in soil.

Chelate enhanced EK remediation is an enhanced EK technology that is used to decontaminate heavy metals-contaminated soil [11,20-21]. The chelate agent can greatly improve the solubility of the heavy metals in soil and maintains their solubility within a wide pH range. This facilitates the removal of heavy metals from the soil. Commonly used chelate agents include ethylenediaminetetraacetic acid (EDTA) [20], ethylenediamine-N,N'-disuccinic acid (EDDS) [22], nitrilotriacetic acid (NTA) [22], diethylenetriamine pentaacetic acid (DTPA), and citric acid [23-24]. Among such chelators, EDTA and DTPA show poor biodegradability, which means that they can easily cause secondary pollution and increase environmental risks. However, the metal complexing ability of NTA and citric acid is poor. Tetrasodium N,N-bis(carboxymethyl) glutamic acid (GLDA) is a new type of biodegradable chelate agent [25-26]. Its complexing capacity for heavy metals is slightly lower than EDTA, but it is a low cost chelator with a low environmental risk. GLDA has been used during the leaching remediation of heavy metal-contaminated soil [27-28]; however, there are currently no reports about GLDA enhancing EK remediation, and thus, the enhancement effects and the migration mechanisms are unclear.

In this study, a heavy metal contaminated soil was taken as the research object. A batch experiment was used to study the extraction effect and the optimal extraction conditions for heavy metals in a soil treated with GLDA. Different EK remediation treatments were used to explore the extent to which GLDA enhanced the electrokinetic migration of heavy metals. This study provides a new option for chelate-enhanced EK remediation.

2. MATERIALS AND METHODS

2.1 Soil and chemicals

Soil was collected from a demolished electroplating factory in Changshu City, Jiangsu Province, China. It was air-dried and ground to pass through a 10 mesh (2.0 mm) sieve. The soil pH,

electroconductivity (EC) (ratio of soil to water = 1:5), organic matter (OM), cation exchange capacity (CEC), and clay content were 5.16, 0.13 mS cm⁻¹, 8.51 g kg⁻¹, 16.8 cmol kg⁻¹, and 32.3%, respectively. The total Cu, Ni, Cr, Pb, Zn, and Cd contents in the soil were 498 mg kg⁻¹, 239 mg kg⁻¹, 652 mg kg⁻¹, 31.9 mg kg⁻¹, 96.7 mg kg⁻¹, and 0.21 mg kg⁻¹, respectively. The Cu, Ni, and Cr contents were above the risk screening values for China national standard GB 15618–2018 (Cu 50 mg kg⁻¹, Ni 60 mg kg⁻¹, and Cr 150 mg kg⁻¹). The European Community Bureau of Reference (BCR) fractions for Cu, Ni, and Cr are shown in Table 1.

Table 1. Heavy metals BCR fractions in the tested soil

Fractions	Cu		Ni		Cr	
	mg kg ⁻¹	percentage	mg kg ⁻¹	percentage	mg kg ⁻¹	percentage
F1	103.5	20.8%	54.5	22.8%	3.10	0.48%
F2	229.6	46.1%	51.8	21.7%	68.0	10.4%
F3	77.8	15.6%	27.4	11.5%	248.1	38.1%
F4	87.1	17.5%	105.3	44.0%	332.8	51.0%
Total	498.0	100%	239.0	100%	652.0	100%

Note: F1: acid soluble fraction, F2: Fe-Mn oxide bound fraction, F3: organically bound fraction, F4: residual fraction.

All the reagents used in this research were analytical grade and were purchased from the Sinopharm Chemical Reagent Company (Shanghai, China). Three types of biodegradable complexing agents were used. These were lactic acid (LA), citric acid (CA), and tetrasodium N,N-bis(carboxymethyl)-L-glutamate (GLDA). The GLDA was purchased from Akzo Nobel Chemicals Co., Ltd. (Shanghai, China). The LA and GLDA solid contents were 90% and 47%, respectively. Deionized water (> 18 MΩ*cm) was obtained from a Milli-Q water system (Millipore Ltd., USA) and was used in all the experiments. The selected anion exchange membrane (AEM) and cation exchange membrane (CEM) were obtained from Tokuyama Soda Ltd., Tokyo, Japan.

2.2 Batch extraction experiments

The concentrations of the three types of biodegradable complexing agents (LA, CA, and GLDA) were all 50 mmol L⁻¹ and the solution pHs were adjusted from 2.5 to 11.5 by adding 0.5 mol L⁻¹ HCl and 0.5 mol L⁻¹ NaOH. The initial 50 mmol L⁻¹ solution pHs for LA, CA, and GLDA were 2.54, 2.28, and 11.9, respectively. The batch extraction experiments were conducted by adding 1.00 g soil to 10 mL solutions with different pHs (solution to soil ratio was 10:1). After shaking at 200 rpm for 2 h (25°C), all samples were centrifuged at 8000 rpm for 5 min and then filtered through 0.45 μm polyethersulfone membrane filters (Nanjing Ronghua Ltd., China). The leachate solution pHs were measured as the equilibrium solution pH, and the Cu, Ni, and Cr concentrations were analyzed by ICP-OES (iCAP 7200,

Thermo Fisher Scientific Ltd., USA). The extraction ratios for the heavy metals were calculated using the following equation:

$$ER (\%) = (V_{\text{solution}}C_{\text{equilibrium}} / Q_{\text{soil}}C_{\text{metal}}) * 100\%$$

where ER (%) is the extraction ratio of the heavy metal, V_{solution} (mL) is the extractant volume, $C_{\text{equilibrium}}$ (mg L⁻¹) is the heavy metal concentration in the equilibrium solution, Q_{soil} is the quantity of soil (g), and C_{metal} is the heavy metal content in the soil (mg kg⁻¹).

2.3 Electrokinetic remediation experiments

2.3.1 Experimental device

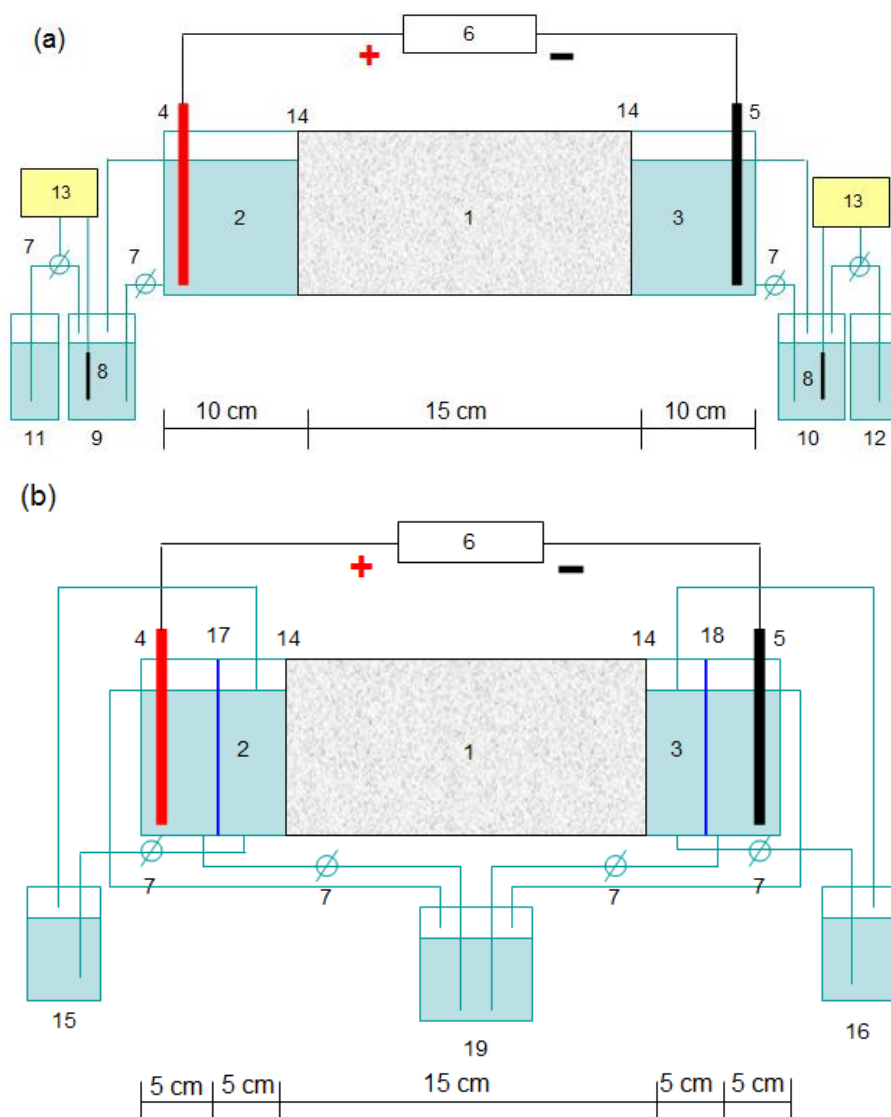


Figure 1. Schematic diagram of electrokinetic remediation. 1, Contaminated soil; 2, Anode pool; 3, Cathode pool; 4, Anode; 5, Cathode; 6, DC power; 7, Peristaltic pump; 8, pH electrode; 9, Anolyte; 10, Catholyte; 11, Acid pool; 12, Alkali pool; 13, pH control director; 14, sintered glass; 15, Anode treating pool; 16, Cathode treating pool; 17, Anion exchange membrane; 18, Cation exchange membrane; 19, Mixing pool.

A schematic diagram of the experimental device is shown in Fig. 1 and is based on Song et al. (2019) [29]. The EK column was a 35 cm (length) \times 8 cm (width) \times 5.5 cm (height) rectangular plexiglass column in which the tested soil compartment (15 cm length) and two electrolytic cells (10 cm length) were separated by sintered glass filters. The electrolytic pools were connected to the solution reservoirs by a silicone tube with a peristaltic multichannel pump at a flow rate of 15 mL min⁻¹.

The electrolytic solution in the two electrolytic pools was flat and there was no external hydraulic gradient. Figure 1(a) shows that the solution reservoirs were connected to a pH control system to control the solution pH needed for each experimental treatment. Figure 1(b) shows that the AEM and CEM were installed in the anode pool and cathode pool, respectively, which were divided into an anode pool/anode concentrator and a cathode pool/cathode concentrator, respectively. The anode and cathode pools were connected by a mixing pool to neutralize the H⁺ and OH⁻ from the anode and cathode. Two square ruthenium oxide coated titanium (Ti/RuO₂) alloy sheets (dimension 3 cm \times 3 cm) were used as electrodes. These were connected to a DC power supply (ITECH, model IT6322A, China) via wires.

2.3.2 Experimental treatments

Table 2 shows that four experiments were conducted, and these were named T1–T4. The EK remediation device shown in Fig. 1(a) was used in T1–T3 and the device in Fig. 1(b) was used in T4. A total of 700 g of soil was added to the column and 0.01 mol L⁻¹ NaNO₃ was used as the electrolyte. The pH of the catholyte in T1 was controlled at pH 4 by 0.5 mol L⁻¹ HNO₃. Exactly 50 mL of 100 mmol L⁻¹ GLDA solution (EC = 13.75 mS cm⁻¹) was added to the soil column by surface addition in T2–T4. The pHs of the anolyte and catholyte in T3 were controlled at pH 6 by 0.5 mol L⁻¹ NaOH and 0.5 mol L⁻¹ HNO₃, respectively. In T4, the AEM and CEM were installed in the anode pool and cathode pool, respectively.

Table 2. Experimental treatments

	Anolyte	Catholyte	GLDA	Voltage gradient
T1	0.01 mol L ⁻¹ NaNO ₃	0.01 mol L ⁻¹ NaNO ₃ (pH was control at 4)	without GLDA	1 V cm ⁻¹
T2	0.01 mol L ⁻¹ NaNO ₃	0.01 mol L ⁻¹ NaNO ₃	5 mmol GLDA	1 V cm ⁻¹
T3	0.01 mol L ⁻¹ NaNO ₃ (pH was control at 4)	0.01 mol L ⁻¹ NaNO ₃ (pH was control at 6)	5 mmol GLDA	1 V cm ⁻¹
T4	0.01 mol L ⁻¹ NaNO ₃ (AEM)	0.01 mol L ⁻¹ NaNO ₃ (CEM)	5 mmol GLDA	1 V cm ⁻¹

Note: AEM, anion exchange membrane; CEM, cation exchange membrane.

The EK treatments lasted for 8 days, during which 1 V cm^{-1} was applied, and the electric currents and electroosmotic flow (EOF) were recorded. The electrolyte solutions in the anode and cathode pools were collected at 0, 6, 24, 48, 96, 144, and 192 hours, and the pH, and Cu, Ni, and Cr concentrations of these solutions were measured. After each experiment, a soil sample was taken and divided into five equal parts, which were named S1, S2, S3, S4, and S5 from the anode to cathode. Then the soil pH, EC, and Cu, Ni, and Cr contents were determined.

2.4 Analytical methods

Electrical current was directly recorded by the automatic acquisition system in the power supply equipment. The EOF was calculated by recording the anode solution volume change. Soil pH, EC, OM, and CEC were analyzed using conventional analytical methods [30]. The soil pH and EC were measured by a pH meter (Shanghai REX Instrument Factory, model pHS-3B, China) and an EC meter (Shanghai REX Instrument Factory, model DDS-11A, China), respectively (ratio of soil to water = 1:2.5). The Cu, Ni, and Cr concentrations in the solutions were analyzed by ICP-OES (iCAP 7200, Thermo Fisher Scientific Ltd., USA). The total Cu, Ni, and Cr contents in the soil were digested in a mixture of HNO_3 - HCl - HF (10:10:1, v/v) using a microwave digestion system (ETHOS-ONE, Milestone, Italy). The digestion solutions were analyzed using ICP-OES (iCAP 7200, Thermo Fisher Scientific Ltd., USA). Blank and standard reference soils (GBW07401 and GBW07406) from the China National Center for Standard Material were used for quality control in the digestion and analysis processes. The Cu, Ni, and Cr chemical fractions in the soils were determined according to the BCR method [31]. There were four fractions: acid form, Fe-Mn oxide bound form, organically bound form, and residual form.

3. RESULTS AND DISCUSSION

3.1 Heavy metal extraction rates by the different reagents

Figure 2 shows the soil heavy metal extraction rates for the different chelating agents under different pH conditions. The Cu and Ni extraction rates by GLDA were the highest, reaching 39% to 48%, followed by CA (26% to 41%) and LA (0.44% to 25%). Wu et al. (2015) studied the effective removal of heavy metals from industrial sludge with the aid of GLDA and found that about 82% Ni and 84% Cu could be effectively extracted at pH 4 [26]. The removal rates were higher than those in our study, which was probably because a higher molar ratio of GLDA: Metal(II) (3:1) was used. The LA extraction rate was the lowest because the complexing constants for LA with the metals were lower than for CA/GLDA. Different solution pHs had different effects on the extraction rates of the three chelating agents. Solution pH had no effect on the GLDA extraction rate, whereas the CA extraction rate was highest at a neutral pH. The solution pH had a considerable influence on the LA extraction rate, which was high under acidic conditions ($\text{pH} < 5$), but decreased rapidly as the solution pH increased.

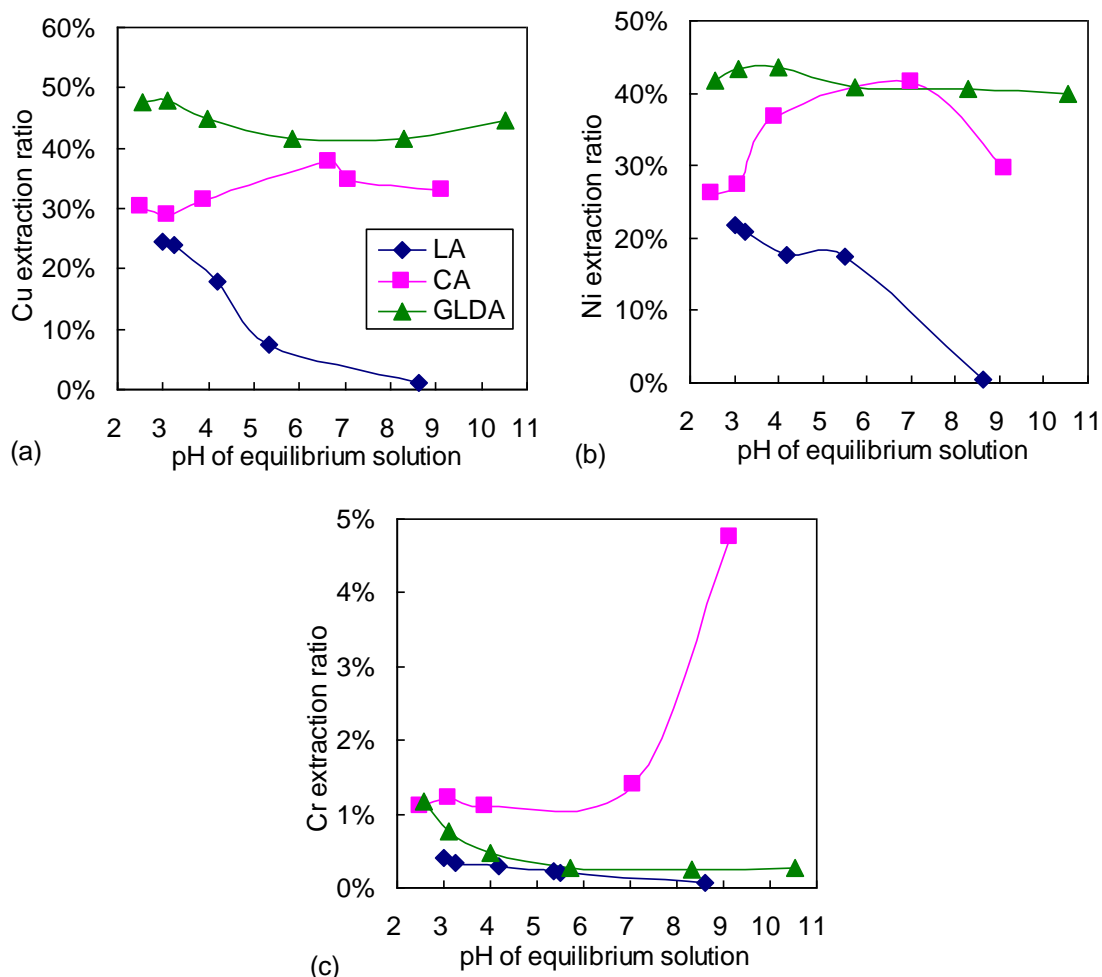


Figure 2. Soil heavy metal extraction ratios at different solution pHs using different chelators (LA: lactic acid; CA: citric acid; GLDA: tetrasodium N,N-bis(carboxymethyl)-L-glutamate).

Figure 2(c) shows that the Cr extraction rates for LA, CA, and GLDA were 0.06%–0.36%, 0.99%–4.28%, and 0.22%–1.07%, respectively. The Cr extraction rates for all the chelating agents were very low and far below the Cu and Ni extraction rates, which may be due to the lower water-to-soil ratio [32] and the different heavy metal fractions in the soil (the Cr residual form percentage was more than 50% and higher than for the Cu and Ni residual forms) (Table 1). Increasing the solution pH slowly reduced the LA and GLDA extraction rates, but the CA extraction rate significantly increased. The reasons for this need further study.

3.2 Changes to electric current and electroosmotic flow

Figure 3(a) shows the changes in electric currents over time. The electric current across the soil column is highly related to the mobile ion concentration in the system [33]. The initial currents in T2–T4 (10–17 mA) were more than in T1 (6 mA) (without GLDA), which was probably because the GLDA solution had a high EC (13.7 mS cm^{-1}). The current in T1 increased gradually and was higher than the currents in T2–T4 after 48 h. This was because acid was continuously added to the system to control the

catholyte pH at 4. The current in T1 increased to 87 mA at the end of EK experiment. The current in T3 was lower than in T1, which might be because the electrolyte pH was controlled to pH 6. The current in T4 decreased gradually to 2 mA and was the lowest of all the treatments. This may be because the AEM in the anode and CEM in the cathode hindered ion transport [15-16].

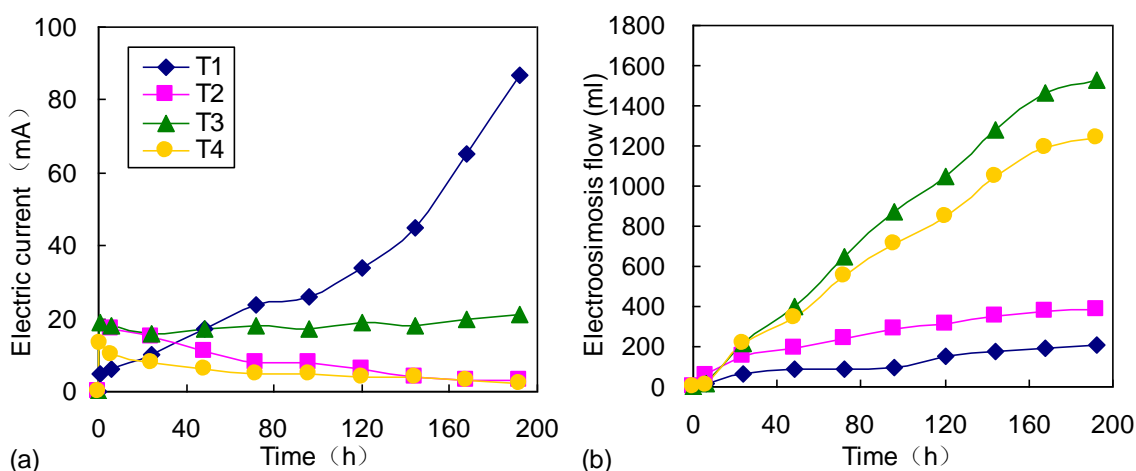


Figure 3. Electric current and electroosmotic flow changes during the treatments (the anode to cathode direction was positive).

Figure 3(b) shows the cumulative EOF changes in the different treatments. The EOF in T3 was the highest among all treatments because the smaller ionic strength and the higher pH led to a thicker diffuse double layer, which was conducive to the generation of EOF [34]. The EOF in T4 was similar to T3, which was attributed to high pH in the T4 system. The EOF for T1 was the lowest in all treatments. Catholyte pH control reduced the soil pH and compressed the diffuse double layer, which inhibited the generation of EOF.

3.3 Changes to heavy metals concentrations in the anolyte and catholyte

As shown in Fig.4, the Cu, Ni, and Cr concentrations in the anolyte and catholyte were significantly different. The Cu and Ni concentrations in the catholyte were higher than in the anolyte. This indicated that Cu and Ni mainly migrated from the anode to the cathode, which showed that the electroosmotic flow velocity of heavy metal chelates was faster than the electromigration velocity. This confirms the results of previous reports. Fan et al. (2014) found that persulfate ions migrated to the cathode and that EOF was the dominant factor affecting the migration of persulfate ions [35]. The Cr concentrations in the catholyte and anolyte were very low (about 0.1 mg L^{-1}), which was consistent with the low removal rate for Cr. The conditions used in this study meant that it was difficult to remove Cr from the soil.

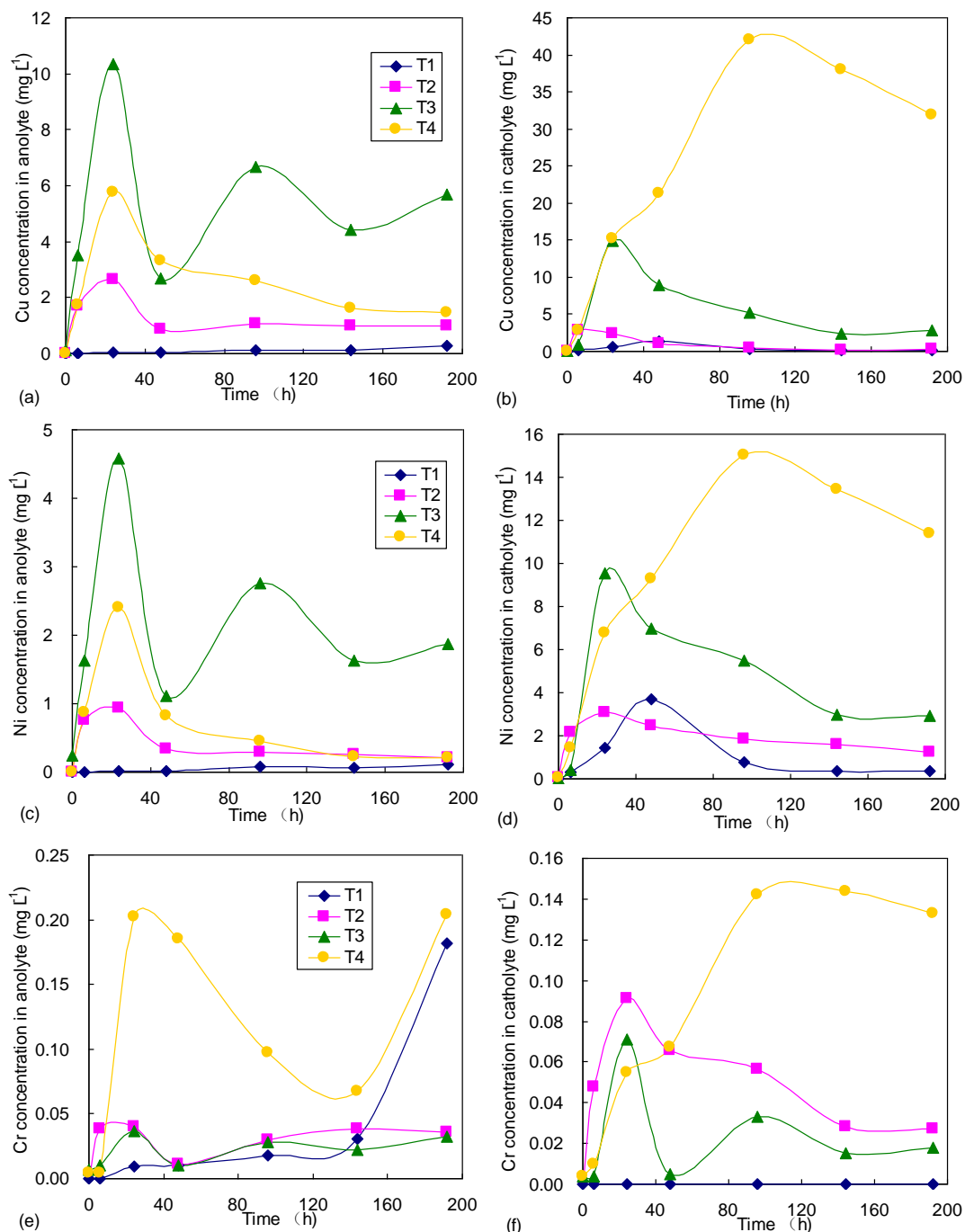


Figure 4. Heavy metal concentration changes in the anolyte and catholyte pools.

The heavy metal concentrations in the different treatments were clearly different. In T1, the Cu concentrations were 0.01–0.26 mg L⁻¹ in the anolyte and 0.01–1.30 mg L⁻¹ in the catholyte, and these values were significantly lower than for the other treatments. The Cu concentrations in the anolyte ranged from 0.01 to 10.3 mg L⁻¹, and were in the order of T3 > T4 > T2 > T1. The Cu concentrations in the catholyte ranged from 0.03 to 41.9 mg L⁻¹, and were in the order of T4 > T3 > T2 > T1. The highest Cu concentration was in T4 where it reached 41.9 mg L⁻¹, which indicated that the ion membrane treatment improved Cu removal from the soil [15,36]. The Ni concentration changes in the different treatments

were similar to Cu, and the highest Ni concentrations in the anolyte and catholyte were 4.58 mg L^{-1} and 15.1 mg L^{-1} , respectively.

The variation in Cr concentration under the different treatments was different from Cu and Ni. In the T4 treatment, the Cr concentrations in the anode and cathode pools were higher than in the other treatments, except for 0–48 h in the cathode pool. However, the Cr concentrations in the electrolytes remained very low throughout the experiment.

3.4 Changes to soil pH and EC distribution after the EK treatments

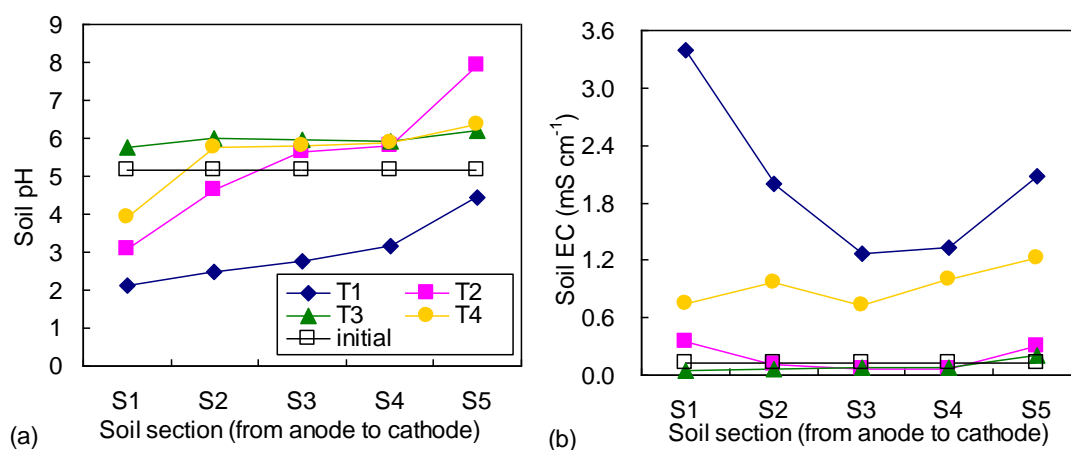


Figure 5. Soil pH and EC changes after the treatments.

The change of soil pH distribution after EK treatments is shown in Fig.5(a). All the soil pHs gradually increased from the anode to the cathode, but the pHs of different treatments varied greatly. The soil pHs for T1 (2.11–4.44) were lower than in the other treatments and the initial soil pH because of the need to control the catholyte pH to 4. Studies had shown that neutralizing the catholyte pH led to a lower soil pH [13]. Therefore, the T1 soil physicochemical properties significantly changed. The anolyte and catholyte pH in T3 were controlled at pH 6. Therefore, the soil pHs varied very little (from 5.76 to 6.21). The changes to soil pH in T4 were similar to T3, but the pH of S1 was 3.94, which may have been caused by polarization of the ionic membranes at the anode [29,37]. The largest soil pH change occurred in T2 (from 3.08 to 7.92), which was similar to previous studies where no pH control measures had been used [14,38].

Figure 5(b) shows that the soil ECs for T1 were the highest ($1.33\text{--}3.40 \text{ mS cm}^{-1}$) of all the treatments and were higher than the initial soil EC (0.13 mS cm^{-1}). Zhou et al. (2005) reported similar results, which they attributed to the addition of acid to control the catholyte pH at 4 [14]. The soil ECs for T3 ($0.05\text{--}0.20 \text{ mS cm}^{-1}$) were close to the initial soil EC, which indicated that T3 (the pHs of the anolytes and catholytes were controlled at pH 6) had little effect on soil properties. The soil ECs for T4 were higher than for T2, T3, and the initial soil EC, which was not consistent with our predictions. However, Song et al. (2019) suggested that the ion exchange membranes prevented ions from migrating out of the soil [29].

3.5 Heavy metal changes in the soils after the EK treatments

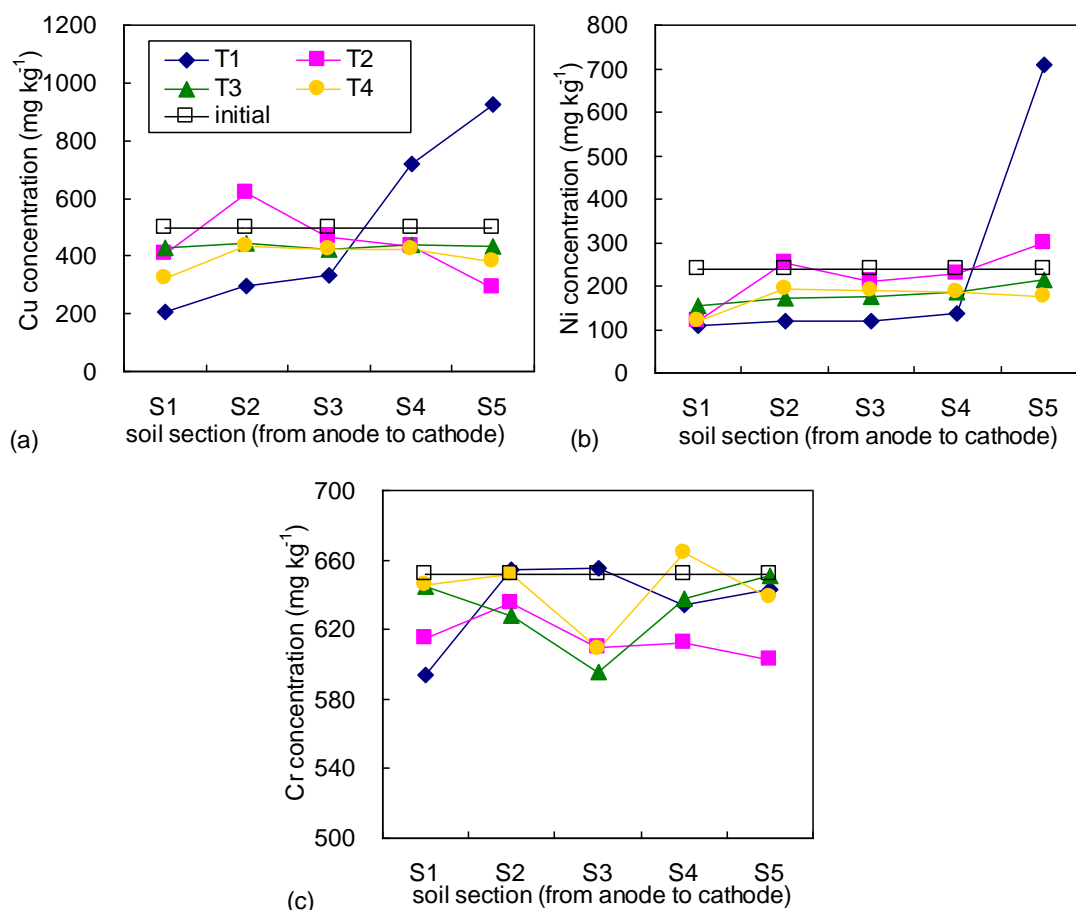


Figure 6. Heavy metal concentration changes in the soil after the treatments.

Figure 6 shows the Cu, Ni, and Cr content changes in the soil after EK remediation. Although the T1 treatment significantly reduced the Cu content in the soil from S1 to S3 (204–336 mg kg⁻¹), a large amount of Cu accumulated at S4 (719 mg kg⁻¹) and S5 (926 mg kg⁻¹), and the total Cu removal rate was only 0.44%. These results indicated that Cu was not removed from the soil, which may be due to the short treatment time (192 h). The total removal rates for T3 and T4 were 12.9% and 20.1%, respectively, and the Cu distribution in each soil section was relatively similar. The Ni distribution in the soil was similar to Cu, and the Ni removal rates were 0.24% (T1), 6.9% (T2), 24.8% (T3), and 27.7% (T4). In T1, the total Ni contents at S1–S4 (108–136 mg kg⁻¹) decreased significantly. However, a large amount of Ni accumulated at S5. This suggests that a longer treatment time was needed. Song et al. (2016) reported that a 52% Cu removal rate and a 41% Ni removal rate were obtained after EDSS treatment [22]. These removal rates were higher than those in our study, because of the longer treatment time (21 d).

The total Cr removal rate was only 1.5%–5.7%, and was significantly lower than for Cu and Ni. The batch experiments showed that the Cr extraction rates by GLDA were only 0.22%–1.07% for the different solution pHs (Fig. 2c). Many researchers have focused on the EK remediation of Cr(VI) contaminated soil and have achieved high Cr(VI) removal rates [13,23,39-40]. Nevertheless, the

remediation of Cr(III) contaminated soil was difficult because of the high Cr residual form percentage (Table 1). Similar to our results, Fan et al. (2015) reported that only a 12.4% Cr removal rate was obtained in an EDTA-enhanced EK remediation experiment (15 d) [41].

3.6 Energy consumption

Table 3. Copper removal rates and energy consumption in the different treatments

	Cu removal rate	Energy consumption (kWh t ⁻¹)	Unit energy consumption per removal rate (kWh t ⁻¹ % ⁻¹)
T1	0.44%	158	3623
T2	11.1%	30.1	27.1
T3	12.9%	75.4	58.5
T4	20.1%	19.3	9.60

The energy consumption in each test was also calculated (Table 3). The power consumption in T1 (158 kWh t⁻¹) was much higher than in the other treatments because the addition of acid led to a very high electrical current (Fig. 3a). The unit power consumption per removal rate in T1 (3623 kWh t⁻¹ %⁻¹) was also higher than all the other treatments, which showed that too much power was consumed. Furthermore, the treatment had the lowest Cu removal rate. In T4, the Cu removal rate was the highest (20.1%) and the unit energy consumption was the lowest (9.60 kWh t⁻¹) of all the treatments. This result demonstrated that installing a CEM and an AEM could increase Cu removal rates and decrease energy consumption in chelate-enhanced EK treatments. Kim et al. (2005) also reported that the 54.5% Pb removal rate in the IEM treatment was higher than in the CK treatment and that the 334 kWh t⁻¹ energy consumption was lower than in the CK treatment [16].

4. CONCLUSIONS

GLDA, a new kind of biodegradable and cheap chelate agent, was used along with EK remediation to remediate a heavy metals-contaminated soil. The results of the batch extraction experiments showed that the Cu and Ni extraction rates for GLDA (39% to 48%) were higher than for citric acid (26% to 41%) and lactic acid (0.44% to 25%), and that the solution pH had little effect on the GLDA extraction rate. The Cr extraction rates by the three types of biodegradable chelate were all low at 0.22% to 4.28%. The EK experiments showed that T1 greatly decreased the soil pHs and increased soil ECs, and the Cu and Ni removal rates (0.24%–0.44%) were the lowest of all the treatments. The T3 and T4 treatments had little effect on soil pH and EC. However, the Cu and Ni removal rates were 12.9%–20.1% and 24.8%–27.7%, respectively, and were higher than for T1 and T2. The total Cr removal rates

in all the treatments were very low at 1.55% to 5.70%, which was probably due to the high percentage of the Cr residual form and the low percentages of the acid soluble and Fe-Mn oxide bound forms. The results for energy consumption indicated that T4 had the lowest energy consumption (19.3 kWh t⁻¹). Therefore, installing a CEM and an AEM could increase Cu and Ni removal rates and decrease energy consumption. Furthermore, GLDA combined with IEM could potentially be used improve the EK remediation of heavy metals contaminated soil.

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CONFLICT OF INTEREST

The authors declare that no competing financial conflicts exist.

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