

Study on Electrokinetic Remediation of Pb-contaminated Saturated Sand

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Electrokinetic remediation is a green remediation technology for contaminated soil, and it has the advantages of low cost, easy operation, minimal secondary pollution, and broad application prospects. In this study, electrokinetic remediation and combined electrokinetic remediation experiments were carried out on Pb(II)-contaminated saturated sand. The distributions of current density (I_d), potential gradient (U_g), pH value, and Pb(II) were analyzed, and the energy consumption (E_c) of the electrokinetic remediation was discussed. The results showed that the removal rate (E_r) of Pb(II) reached 98.37% after 24 h when the 0.2 mol/L of KCl was the electrolyte and the U_g was 3 V/cm. With the electrokinetic remediation method, the coated titanium electrode had a significant advantage over the graphite electrode. The electrolysis efficiency increased as the concentration of KCl electrolyte increased. In addition, the conductivity of the KNO₃ electrolyte was higher than that of the KCl electrolyte. During the electrokinetic remediation process, acidification/alkalinization and a focusing effect might occur in the sand samples, which could affect the efficiency of the remediation. The acidification/alkalinization issue could be effectively addressed by adopting the combined electrokinetic remediation method using citric acid or sodium citrate as a buffer solution. The polarity exchange method had little influence on the electrokinetic remediation.

Keywords: electrokinetic remediation; lead contamination; saturated sand; electrolyte; buffer solution

1. INTRODUCTION

With the rapid development of China's industrial technology, the problem of heavy metal pollution has become increasingly serious. According to the "National Survey Bulletin on Soil

Pollution”, issued by the Ministry of Environmental Protection and the Ministry of Land and Resources of China, in 2014, a total of 16.1% of the soil in China contain more than the standard amount of heavy metals. The over-standard rate in cultivated land is 19.4%, the over-standard rate of heavy metal related enterprise and their surrounding soils is 36.3%, and the over-standard rate of solid waste treatment plants is 21.3%[1]. The soil quality is inferior, and the pollution problem is prominent in China, which has seriously threatened the safety of people’s lives and properties. Therefore, research on heavy metal contaminated soil is particularly important.

As one of the three heavy metal pollutants that affect hormones, Pb has long been used in industrial products, such as batteries, dyes, building materials, and gasoline. It is widely distributed in soil, water, air, and other environments[2,3]. Pb can enter the animal and human body through the food chain and the drinking water, which in turn affects human health and the ecological environment[4-6]. Common remediation methods for heavy metal pollution include bioremediation, chemical remediation, and physical remediation[7-11]. The electrokinetic remediation method has attracted extensive attention because of its high efficiency, low cost, and minor secondary pollution[12]. Suzuki *et al.*[13] investigated the influence of the ethylene diaminedisuccinic acid (EDDS) concentration, the potential gradient (U_g), and the electrode materials on the removal efficiencies of Pb and Cd from soils. It was found that the Pb removal rate (E_r) was enhanced by increasing either the voltage or the EDDS concentration. The platinum-coated titanium electrode and iridium dioxide-coated titanium electrode also performed better than the graphite electrode. Hanay *et al.*[14] analyzed the E_r and migration ability of Cr, Pb, and Zn in sewage sludge using ethylene diaminetetraacetic acid (EDTA) as the washing solution and applying the sequential chemical extraction scheme according to the Community Bureau of Reference (CBR) guidelines. The highest E_r was 34% for Cr, 27% for Pb and 20% for Zn. The migration ability was found to follow the order of Cr>Pb>Zn. Shrestha *et al.*[15] conducted an electrokinetic remediation study on the sediment from the WeisseElster River, Germany. A columnar experimental assembly was employed, and the voltage was set at 3 V. The results showed that the heavy metal ions Cd, Pb, and Zn migrated from anode to cathode under the applied electric field. Then, these metals precipitated at the cathode, and finally the metal concentrations decreased to zero. Wan *et al.*[16] studied the effects of the U_g , the remediation time, the electrolyte type, and the electrified mode on the electrokinetic remediation of Pb-contaminated soil by preparing Pb-contaminated soil and building an electrokinetic remediation test device. Ren *et al.*[17] improved the effect of remediation by adding an activated carbon fiber and cation membrane between the soil chamber and the cathode chamber. The effect of the pH of the citric acid-sodium citrate anolyte on the electrokinetic remediation of Pb-contaminated soil was studied in an electric field of 1 V/cm on an artificially configured Pb-contaminated soil. The results showed that when Fe(III)-activated carbon fiber was used to enhance the electrokinetic remediation; the smaller the anolyte pH, the better the removal of Pb. Zhang *et al.*[18] used ascorbic acid and citric acid as enhancing agents and investigated their effects on the electrokinetic remediation of Pb-contaminated tailing soil with high acid buffer capacity. The effects of ascorbic acid (as a saturated liquid in the soil) and the U_g on the electrokinetic remediation of Pb-contaminated soil was systematically analyzed using 0.1 mol/L citric acid as the catholyte with a pH of 2-3. When ascorbic acid was 0.4 mol/L at the cathode chamber with a pH of 2-3 (controlled by citric acid) and an applied U_g of 2 V/cm, the migration ability of Pb in soil was

significantly improved, achieving a good remediation effect. Cai *et al.*[19] studied the electrokinetic remediation of Pb-contaminated soil in the Pearl River Delta region, China, using a polarity exchange technique to improve the conventional electrokinetic remediation of Pb-contaminated soil. The relations between the E_r of Pb and the changes in soil pH, the treatment time (t), and the frequency of polarity exchange were investigated. When the U_g was 1 V/cm and the exchange polarity interval was 48 h, the E_r of Pb was found to achieve a maximum of 87.7%, which was much higher than that obtained by the conventional electrokinetic remediation method (61.8%). Yang *et al.*[20] studied the electrokinetic remediation of Pb-contaminated soil under different remediation times. The distributions of the electrical current, the electrical conductivity, the pH, and the Pb concentration were analyzed. It was found that increasing the t improved the E_r of Pb.

According to the above studies, current studies on electrokinetic remediation mainly focuses on Pb removal from soils. There is little research regarding Pb removal from sands. In this study, electrokinetic remediation and combined electrokinetic remediation were performed on Pb(II)-contaminated saturated sand. The distributions of the current, the potential, the pH, and the Pb(II) concentration in the sand samples were analyzed and compared, and the energy consumption during the electrokinetic remediation was also discussed.

2. EXPERIMENTAL

2.1. Test materials and equipment

The test sand was collected from the local river in Dalian, China. The original sample was sieved by a 1-mm sieve, washed with water, and sieved by a 0.075-mm sieve. The silt and clay were removed from the sand sample. Then, the sand sample was dried and subjected to screening. The non uniform coefficient C_u of the sand is 1.91, the curvature coefficient C_c is 0.89, and the average particle diameter D_{50} is 0.31 mm. The determined maximum and minimum dry densities of the sand are 1.69 and 1.30 g/cm³, respectively.

The test equipment mainly consists of a reaction chamber, a direct current (DC) stabilized power supply, a voltage module, a current module, a solution-replenishing device, an overflow-collecting device, and a data collection system, as shown in Figure 1. The tests were performed using either a titanium electrode or a graphite electrode. The area of the electrode was 100×70 mm². The reaction chamber was constructed of polyvinyl chloride (PVC) plates, with a size of 310×90×120 mm³. The effective size of the soil sample chamber was 100×70×100 mm³ and the effective size of the electrolysis chamber was 70×70×100 mm³. The soil chamber was evenly divided into 10 zones, namely T1, T2, T3, T4, T5, T6, T7, T8, T9 and T10, respectively. The soil chamber and the electrolysis chamber were separated by filter paper, a glass fiber membrane filter, and a porous PVC plate to prevent soil particles from entering the electrolysis chamber. A solution-replenishing hole was placed at the bottom of the cathode chamber and an overflow hole was placed on the sidewall. The electrolyte was pumped into the electrolysis chamber at a constant rate by a peristaltic pump. The

excess electrolyte was discharged from the overflow hole to eliminate the influence of the hydraulic pressure difference on the test results.

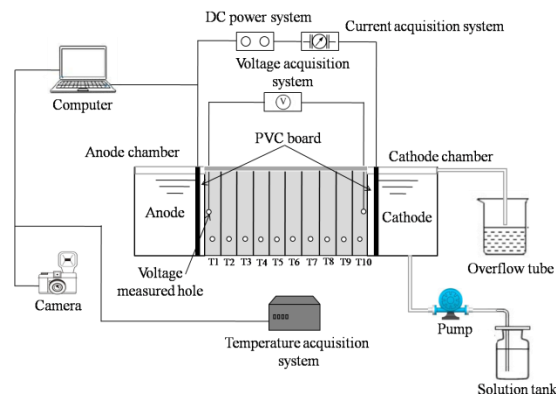


Figure 1. Electrokinetic remediation equipment with DC power system, data collection system, reaction chamber, solution-replenishing device and overflow-collecting device.

2.2. Experiment method

A total of 1.5985 g of PbNO_3 was weighed and dissolved in 1 L of distilled water to obtain a stock solution with a Pb(II) concentration of 1000 mg/L. The Pb(II) solution used in the test was obtained by diluting the stock solution. The PbNO_3 stock solution was first diluted at a ratio of 1:4 to 200 mg/L. A total of 943 mL of the diluted solution was added to 942.88 g of sand. The mixture was stirred to obtain a sand sample with a lead concentration of 200 mg/kg. The sand sample was then transferred to an oven at 105 °C for drying.

Table 1. Test scheme for the electrokinetic remediation and combined electrokinetic remediation

Test	Electrode	U_g (V/cm)	Electrolyte	Polarity exchange
S1	Coated titanium alloy	1	Tap water	---
S2	Coated titanium alloy	3	Tap water	---
S3	Coated titanium alloy	3	0.1 mol/L KCl solution	---
S4	Coated titanium alloy	3	0.2 mol/L KCl solution	---
S5	Coated titanium alloy	3	0.2 mol/L KNO_3 solution	---
S6	Graphite	3	Tap water	---
S7	Coated titanium alloy	3	pH=4.8 citric acid-sodium citrate solution	---
S8	Coated titanium alloy	3	pH=4.8 citric acid-sodium citrate solution 0.2 mol/L KNO_3 solution	---
S9	Coated titanium alloy	3	0.2 mol/L KNO_3 solution	4 h
S10	Coated titanium alloy	3	0.2 mol/L KNO_3 solution	6 h

The measured lead concentration of the sand sample after drying was 193.1 mg/kg. During sample loading, the separator and filter paper were first installed in the test device. Then, the sand sample was added to the reaction chamber in 5 lots and compacted to a compactness of 0.7 after loading. The electrolyte was added to the anode and cathode electrolysis chambers simultaneously to ensure no hydraulic head difference that could lead to the flow of the solution. The solution was aged for 30 min before the remediation test started. After the test started, the pH values of the solution in the anode and cathode chambers were measured and recorded at 1 min, 5 min, 10 min, 20 min, 40 min, and 60 min, and then recorded every 1 h thereafter. The hydraulic head of the electrolyte was kept constant during the test without solution replenishing or overflow discharge. The electrolyte used in the combined remediation test was a mixture of KNO₃ solution and citric acid-sodium citrate solution (pH=4.8). During the experiment, the hydraulic head of the electrolyte in the anode chamber was kept constant using a Mariotte bottle. The electrolyte solution from the replenishing reservoir was pumped to the cathode chamber at a constant rate by the peristaltic pump. The pumping rate was 0.8 mL/min. The test scheme for the electrokinetic remediation (S1, S2, S3, S4, S5 and S6) and combined electrokinetic remediation (S7, S8, S9 and S10) is summarized in Table 1.

2.3. Calculation of removal rate and energy consumption

The Pb(II) removal rate (E_r) in the electrokinetic remediation test was calculated as follows:

$$E_r = \frac{(m_0 - m_e)}{m_0} \times 100\% \quad (1)$$

where E_r is the removal rate, %; m_0 is the initial mass of pollutant, mg; and m_e is the mass of the residual pollutant, mg.

The energy consumption (E_c) in the electrokinetic remediation test was calculated as follows:

$$E_c = \frac{1}{M_c} \int UI dt \quad (2)$$

where E_c is the energy consumption to remove a unit mass of pollutant, W·h/mg; M_c is the total removal of pollutant, $M_c = m_0 - m_e$, mg; U is the potential at time t , V; I is the current at time t , A; and t is the treatment time, h.

In order to analyze the change laws of current and potential, we define I_d as current density, mA/cm², and U_g as potential gradient, V/cm.

3. RESULTS AND DISCUSSION

3.1. Distribution of current and potential in the sand during electrokinetic remediation

A total of 6 electrokinetic remediation tests were carried out. The applied U was 10 V for S1 and 30 V for the other five tests. The t was 24 h for each test. The changes in current and potential with time are plotted in Figure 2. It can be seen from Figure 2 that the current distribution and the potential distribution in S4 and S5 were different, even though both tests used 0.2 mol/L of electrolyte. This was caused by different properties of the electrolyte (S4 used KCl, S5 used KNO₃). When KNO₃ was used

as an electrolyte, a large current was formed in the sand sample at the initial stage of the electrolysis, which indicates that the conductivity of the sand sample was enhanced. The I_d achieved its maximum at 6 h of electrolysis time, then gradually decreased and remained constant. The I_d on S4 exhibited a cyclic increase and decrease, which suggests that the conductivity of the KNO_3 electrolyte was higher than that of the KCl electrolyte. The I_d trend of the S3 was similar to that of the S4, but the value of the S3 was smaller than that of the S4. Therefore, for the KCl electrolyte, high concentration was favorable for electrolysis. The electrolytes used in test S1, S2, and S6 were all tap water. The current in the sand samples was small during the electrolysis, which was mainly caused by the poor conductivity of water. The concentration of mobile ions in the pore fluid of the sand is relatively high in the early stage of remediation, and the H^+ produced by the water ionization in the anode electrode chamber produced an acid migration zone moving towards the cathode, which dissolved and desorbed more mobile ions on the surface of the sand during the migration process. Under the action of the directional transfer, a higher current was generated[21,22]. When most of the mobile ions move out of the sand, the current decreased accordingly[23,24]. It can be seen from Figure 2(b) that the U_g in the sand increased with the increase of electrolysis time. The U_g increases in S3-S5 were greater than those in S1 and S2, and the increase in S6 was the smallest. The increase in U_g indicates that as the electrolysis progressed, the average resistance of the sand sample increased gradually, and the electrolysis efficiency continuously decreased.

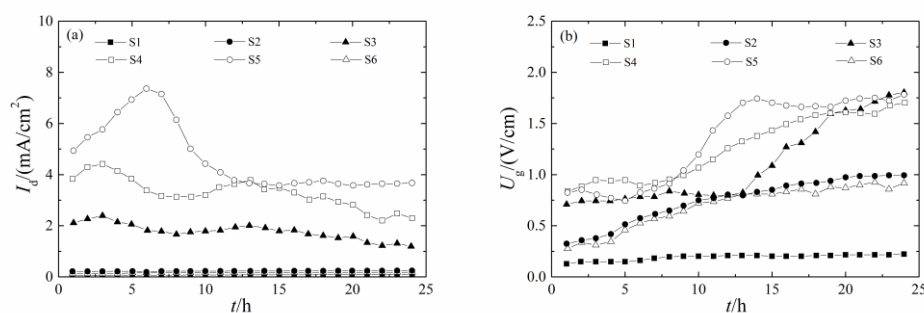


Figure 2. Electrokinetic remediation tests of S1, S2, S3, S4, S5 and S6 (a) Curves of current density versus treatment time, (b) Curves of potential gradient versus treatment time.

3.2. Distribution of pH in the sand during electrokinetic remediation

The change in sand pH during the electrokinetic remediation is shown in Figure 3. The initial pH of the sand sample was 5.61. The sand pH increased from the anode side to the cathode side, and the range of the acidic zone was larger than that of the alkaline zone. The water in the anode chamber was oxidized to generate hydrogen ions and oxygen, and the water in the cathode chamber generated hydrogen ions and oxygen. With the progress of electrokinetic remediation, the anode produced a lot of H^+ , and lead to the decreased of the pH to form an acidic zone. Under the action of electric field, H^+ migrated to the cathode made the acidic zone moved to the cathode. On the contrary, the cathode produces a lot of OH^- to form an alkaline zone. As the acidic zone, the alkaline zone moved to the

anode under the action of electric field[20, 25-27]. An abrupt change in pH was observed in the T4-T8 zone, which resulted in acidification or alkalization of the sand sample and further led to a decline of the I_d . The current decline was unfavorable for the removal of heavy metal ions. The amount of residual heavy metals in the pH changing region was significantly higher than that in other regions. This phenomenon is called the focusing effect[28]. After the test was completed, the sand pH in S2-S6 changed significantly under the influence of electrolysis. The change rate and change amount of the sand pH in S3-S5 (with a strong electrolyte) were much larger than those in S1, S2, and S6. After the test was completed, the pH of the sand near the cathode was higher than 8, and even higher than 10 for S3-S5. At the same time, the pH of the sand near the anode was significantly reduced to 2-4. However, the sand pH in S1 did not change significantly. This was mainly caused by the poor conductivity of the electrolyte, the low U_g , and the slow electrolysis reaction process.

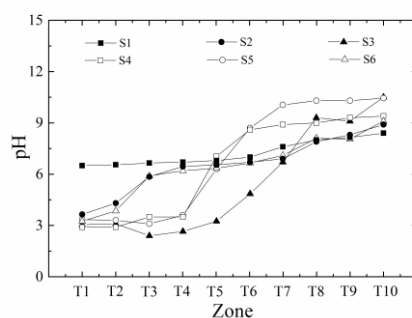


Figure 3. pH distribution curves of electrokinetic remediation tests of S1, S2, S3, S4, S5 and S6.

3.3. Distribution of Pb(II) in the sand after electrokinetic remediation

To analyze the effect of electrokinetic remediation, the sand samples were sliced and tested to determine the distribution of Pb(II) in the samples. The results are shown in Figure 4. The distributions of residual Pb were basically the same except for S6. The concentration of Pb(II) was lower near the anode and higher near the cathode. The main reason is that the pH of sand samples closer to the anode was the lowest during the remediation process, which more conducive to the removal of Pb (II). Whereas, the Pb(II) in the cathode and the hydroxide ions formed a difficult dissolved substance, which impeded the migration of Pb(II) to a certain extent[29]. Since S1 used tap water as the electrolyte and the potential was 10 V, the E_r of Pb(II) was poor and more residue was found near the cathode. S6 used a graphite electrode, which caused low electrolysis efficiency and the worst E_r of Pb. S3 and S4 used 0.1 mol/L and 0.2 mol/L KCl as electrolytes, respectively. The E_r of Pb were basically the same for S3 and S4. In general, the E_r of Pb of S1-S5 were relatively high, and E_r of Pb reached 98.37% for S3. Therefore, when using the electrokinetic remediation method for Pb removal, a good removal effect can be achieved by using coated titanium alloy as an electrode, a U_g of 3V/cm, 0.1 mol/L KCl as an electrolyte, and a t of 24 h.

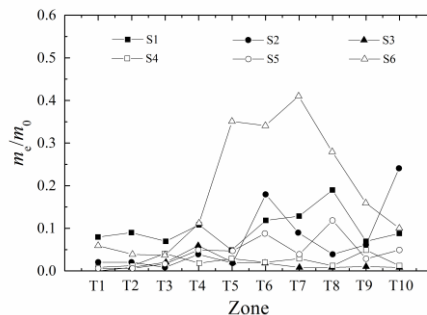


Figure 4. Pb(II) distribution curves of electrokinetic remediation tests of S1, S2, S3, S4, S5 and S6.

3.4. Distributions of current and potential in the sand during combined electrokinetic remediation

The above analysis showed that the electrokinetic remediation method can effectively remove the Pb contamination from the sand samples. However, it was found that the electrode is prone to polarization and a focusing effect can occur in the sand sample, which greatly limits the electrokinetic remediation efficiency[30]. Therefore, the polarity exchange technique was adopted for further analysis. In Figure 5, the changes in current and potential in S9 and S10 are shown. When the electrode polarity was reversed, the I_d and U_g might drop or rise with a wave-shaped distribution. The main reason is that the H^+ and OH^- in the original cathode and anode chambers underwent a neutralization reaction in the electrolysis chamber after reversing the electrode polarity. Thus, the charge density in the electrolyte and in the sand sample decreased. Since the I_d was proportional to the mass fraction of the ions in the sand, the I_d and U_g also dropped. The E_r of Pb(II) was low in S9 and S10. This is because when the electrode polarity was reversed; the Pb(II) precipitated in the cathode chamber was converted to the ion form and re-entered the sand sample, which resulted in the increased of current value[16].

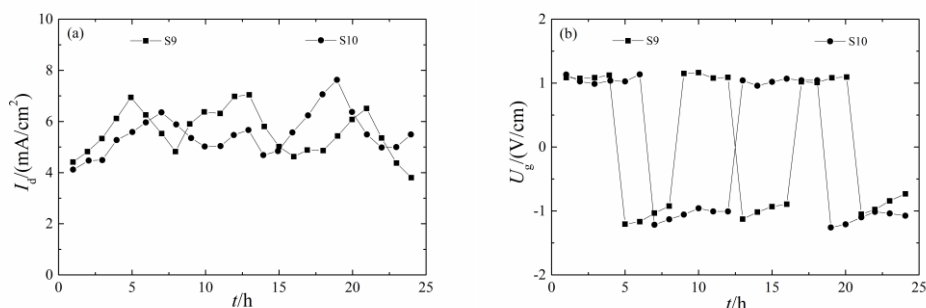


Figure 5. Combined electrokinetic remediation tests of S9 and S10 (a) Curves of current density versus treatment time, (b) Curves of potential gradient versus treatment time.

3.5. Distribution of sand pH during combined electrokinetic remediation

To address the acidification/alkalinization issue, a citric acid-sodium citrate buffer solution with pH=4.8 was added to the electrolyte.

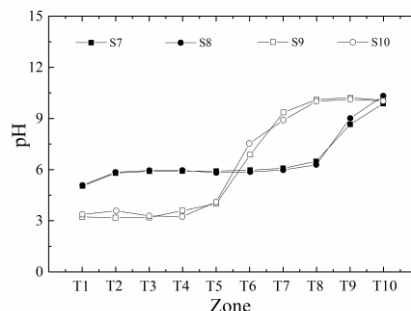


Figure 6. pH distribution curves of combined electrokinetic remediation tests of S7, S8, S9 and S10.

The electrode polarity was periodically exchanged to improve the efficiency of the electrokinetic remediation. In Figure 6, the distribution of sand pH during the combined electrokinetic remediation is shown. When citric acid was used as the electrolyte or a buffer (S7 and S8), a large pH change was observed only in regions adjacent to the cathode (T9 and T10). The pH changes in other regions were small. This result indicates that using citric acid as a buffer can effectively solve the acidification/alkalinization problems during the electrolysis and improve the efficiency of electrokinetic remediation. When the reaction chamber kept in acid environment, it is beneficial to resolve of Pb(II) and promote the migration of Pb(II), which improved the E_r [31]. S9 and S10 did not use citric acid as a buffer but only exchanged the electrode polarity at a constant frequency. The acidification/alkalinization problem also appeared in the S9 and S10 samples.

3.6. Pb(II) distribution in the sand after combined electrokinetic remediation

In Figure 7, the distribution of Pb in the sand samples after the combined electrokinetic remediation is shown. The data show that the Pb removal effect of the combined electrokinetic remediation method was poor. In the S7 and S8 tests, Pb(II) could form different ligands with citric acid and sodium citrate. The ligands could have various geometries and structures[32]. The coordination number of the ligands could be 2-10[33]. Moreover, because of the difference in the pH of the complexation reaction, the complexes that could be formed between Pb(II) and citric acid or sodium citrate include $Pbcit$, $PbHcit$, $Pbcit_2$, Pbh_2cit_3 , PbH_4cit_2 , $Pb(OH)_2cit_2$, $Pb_2(OH)_3cit$ and others (where $cit=C_6H_5O_7^{3-}$)[34]. The porous structure of sand caused enrichment of the complex. The lead ion complex could not be discharged by the electric field, which resulted in a decrease in the E_r .

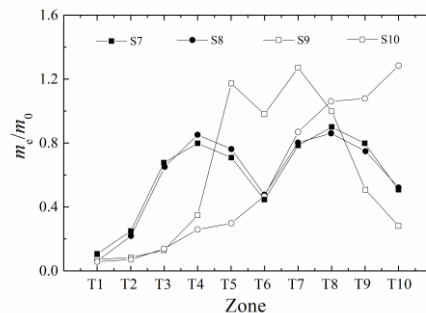


Figure 7. Pb(II) distribution curves of combined electrokinetic remediation tests of S7, S8, S9 and S10.

3.7. Energy consumption analysis of electrokinetic remediation

Table 2 contains the E_r of Pb(II) and E_c in the electrokinetic remediation (S1, S2, S3, S4, S5 and S6) and combined electrokinetic remediation (S7, S8, S9 and S10). When the U_g was high and the electrolyte was strong, the E_r and the E_c were high (S3, S4, and S5), which consistent with the results of reference[35]. In addition, using the KCl electrolyte resulted in a higher E_r and lower E_c than using the KNO_3 electrolyte; therefore, the KCL electrolyte has better economic benefits. However, using KCl as an electrolyte produced a large amount of the toxic byproduct Cl_2 , which is a pollutant in the environment and is therefore not recommended. Methods such as polarity exchange and the addition of a buffer not only reduced the E_r of Pb but also increased the E_c .

Table 2. Removal rate and energy consumption of electrokinetic remediation and combined electrokinetic remediation

Test	E_r (%)	$UI dt$ (W·h)	E_c (W·h/mg)
S1	90.22	2.00	0.01
S2	93.15	21.62	0.12
S3	98.37	278.24	1.41
S4	97.72	607.79	3.11
S5	95.35	872.61	4.58
S6	81.91	17.95	0.20
S7	42.22	220.29	1.34
S8	42.46	733.78	8.69
S9	43.56	883.57	10.40
S10	45.92	890.55	10.22

4. CONCLUSIONS

In this study, the electrokinetic remediation for Pb(II) removal from contaminated saturated sand was investigated. The effects of the current, the potential, the electrode material, the electrolyte, polarity exchange, and a buffer solution were analyzed. The results led to the following conclusions:

(1) The coated titanium electrode had advantages over the graphite electrode. Higher electrolysis efficiency and higher E_r of Pb(II) were achieved with the coated titanium electrode. The electrolysis efficiency increased as the concentration of KCl electrolyte increased. The conductivity of the KNO_3 electrolyte was higher than that of the KCl electrolyte. During the electrokinetic remediation process, acidification/alkalinization and a focusing effect could occur in the sand sample, which could affect the efficiency of the electrokinetic remediation.

(2) When the U_g was 3 V/cm and the electrolyte was 0.2 mol/L KCl, the E_r of Pb(II) reached 98.37% after 24 h of treatment. When using 0.2 mol/L KNO_3 as an electrolyte, the E_r of Pb(II) could achieve 95.35%, which was similar to the KCl case. Considering that using KCl as electrolyte could produce a large amount of toxic Cl_2 , which is unfavorable to the environment, we recommend using KNO_3 as the electrolyte.

(3) When citric acid or sodium citrate was used as a buffer solution, the acidification/alkalinization issue in the sand sample was effectively addressed but the E_r of Pb(II) was not improved. The polarity exchange technique had little effect on the electrokinetic remediation and is therefore not recommended.

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CONFLICT OF INTEREST

On behalf of all authors, the corresponding author states that there is no conflict of interest.

DATA AVAILABILITY

The data used to support the findings of this study are available from the corresponding author upon request.

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