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Electrokinetics removal of lead from lead-contaminated red soils①

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Abstract: Ex-situ electroremediation tests were conducted on the lead-contaminated red soils to find out the optimum condition for the most efficient removal of lead pollution from the red soil, and to examine the relation of the pH of the soil with the electroremediation efficiency. The results show that the electroremediation technology is efficient to remedy Pb contaminated red soils, and the removal efficiency can be enhanced by controlling pH value in the cathode reservoir with HNO_3. The average removal efficiency of Pb is enhanced from 24.5% to 79.5%, and the energy consumption reaches 285 kW·h per m^3 red soil.

Key words: electroremediation; red soil; lead pollution
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1 INTRODUCTION

Applications of electrokinetics phenomena to remove heavy metals from soils have been studied in recent years as one of the promising technologies. This method has certain advantages over other technologies, e.g. it can be practiced in situ, and it is not difficult to implement in the fine textured soils with low hydraulic conductivity. Using this technology, some field trial and cleanup projects have removed toxic heavy metals from industrially polluted soils[1-3]. The theory and principles of the technology have been extensively discussed[1, 4-6].

Despite the good results obtained with a few applications of the electroremediation and some pilot scale tests, the technology is not yet fully mature. Most of the previous laboratory studies were limited to the experiments with commercially available soils such as kaolinite and montmorillonite[7-11]. This study, however, presents the results of an ex-situ test of the electroremediation of the red soils. The objectives of the study are: to explore the flexibility of the electroremediation technology on the removal of heavy metals from red soils, to find out the optimum condition for the most efficient removal, and to examine the relation of the pH value of soil with the electroremediation efficiency.

2 MATERIALS AND METHODS

The samples of the red soils for this study were gathered from a building site located in Yuelu Mountain, Changsha, China. The red soils were stored at 4 ℃ for a week before analysis, then spiked with $Pb(NO_3)_2(s)$, and crashed with a ball mill. The mass ratio of Pb to dry soil of the prepared sample was $(15-21)\times 10^{-3}$. Some of its more important properties were analyzed and the results are listed in Table 1. The red soils had been saturated by the distilled water before remediation. Two 8 μm filter papers were placed at each side of the soil sample to separate the soil cell from the electrode compartment and the electrolyte reservoirs.

Table 1 Chemical and physical characteristics of red soils

pH	w(pb)/%	Cation exchange capability/ (10^{-2}mol·kg^{-1})	$w(H_2O)$/%	w(Carbonate)/%
5.70	0.056	9.16	37.55	4.58

A schematic diagram of the experimental apparatus used in the study is shown in Fig. 1. The size of the experimental apparatus is 400 mm long, 200 mm wide and 200 mm high. The experimental apparatus consists of four main parts: the soil cell, the electrode compartment, the electrolyte reservoirs and the power supply. The system was open. The electrolyte reservoirs were filled with distilled water. The surface level of the water was a few millimeters lower than that of the red soils. A DC power source provided a constant current. In order to find out the optimum condition for the most efficient removal, a pair of control experiments has been conducted. One was on the unenhanced condition; the other was on the enhanced cathode condition.

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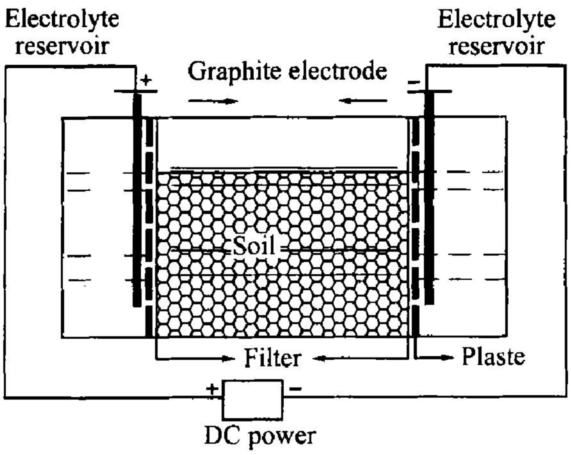

Fig. 1 Schematic diagram of experimental apparatus

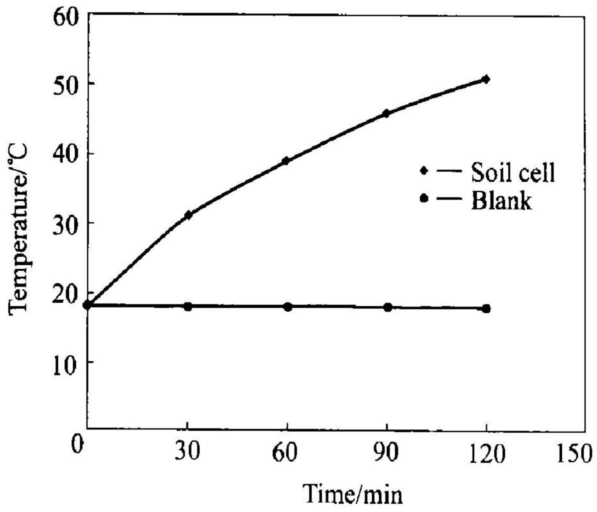

Fig. 2 Relationship between temperature of soil cell and electroremediation time

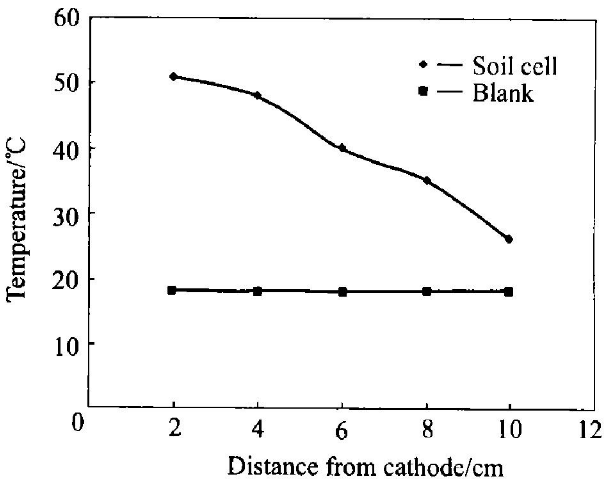

Fig. 3 Variation of temperature of soil cell after 120 min

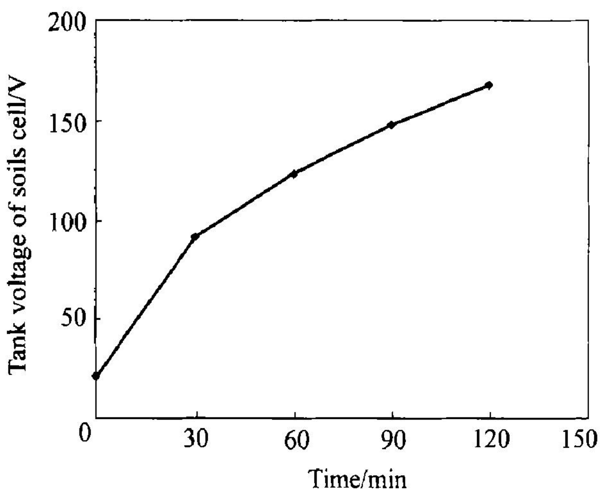

Fig. 4 Tank voltage of soil cell versus electroremediation time

3 RESULTS AND DISCUSSION

The results of two electroremediation tests are listed in Table 2.

Table 2 Comparison of unenhancement test(Test Ⅰ) and cathode acidification enhancement test (Test Ⅱ)

Test	Current density/ $(A\cdot m^{-2})$	Cathode condition control	w(Pb)/ %	Extracted efficiency/ %	Energy consumption/ $(kJ\cdot kg^{-3})$
Test Ⅰ	66.7	Uncontrolled	2.105	24.5	-
Test Ⅱ	33.3	Control with 1 mol/L HNO_3	1.554	79.5	285

3.1 Unenhancement test (Test Ⅰ)

Test Ⅰ is a trial test. Its purpose is to verify the flexibility of the electroremediation technology to remove Pb from the contaminant red soil.

In this test, the temperature of the soil cell close to the cathode increases obviously. The temperature reaches 51 ℃ after 120 min as shown in Fig. 2. The temperature of soil cell is varied gradually after 120 min. The temperature of soil cell decreases as the distance away from the cathode increases as shown in Fig. 3.

Meanwhile the tank voltage of the soil cell rises sharply as shown in Fig. 4. Then the output power of DC power supply is exceeded and the test has to halt. This result accords with the conclusions of Reed[13]: when the current density exceeds 50 A/m^2, temperature effects will become an important factor.

In electroremediation, the application of direct electric current via electrodes immerses in the red soil results in the oxidation of water at the anode, generating an acid front that moves toward the cathode and causes dissolution of metals compound, while the reduction of water at the cathode generating a base front that moves toward the anode and causes precipitation of metals ions. Because the ion mobility of H^+ (36.25 $m^2/(V\cdot s)$) is faster than that of OH^- (20.58 $m^2/(V\cdot s)$), two fronts meet at 7 - 8 cm from the anode as shown in Fig. 5.

After 2 h electroremediation, the Pb content of the soil cell is increased as the distance from the anode increases. The average Pb removal efficiency is 24%, and Pb content of soil at the cell of 0 − 2 cm distance from the anode is the lowest as shown in Fig. 6. The results indicate that the electroremediation technology is applicable to removal Pb contaminants from the red soils; and electroremediation effect starts from the nearest soil cells to the anode.

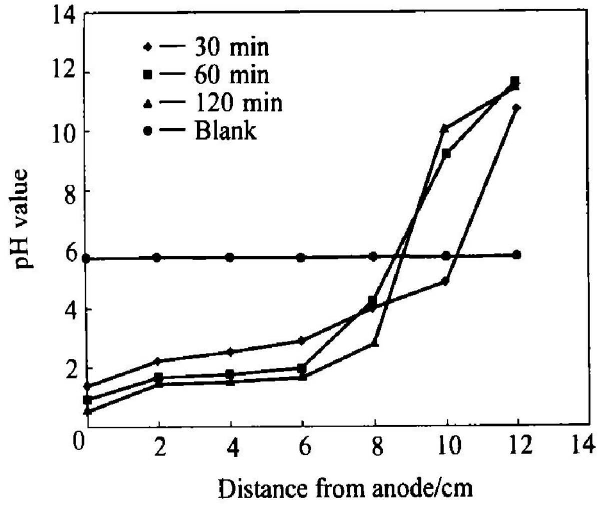

Fig. 5 pH value versus distance from anode

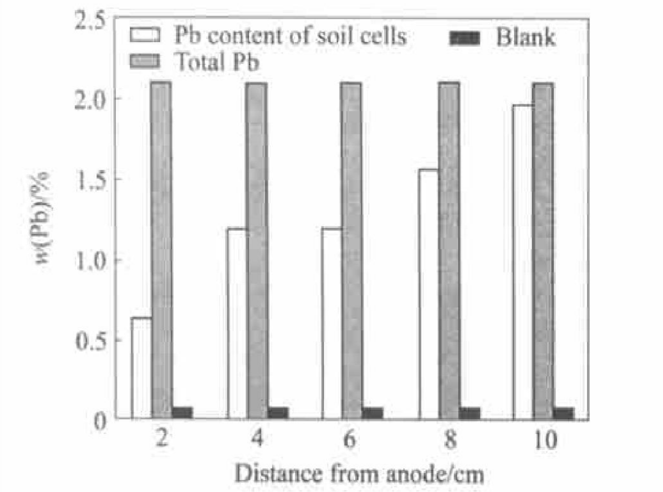

Fig. 6 Migration of Pb after 2 h

3.2 Cathode acidification enhancement test (Test II)

Test II is based on test I. In order to improve the electric conductivity of the soil cells close to the cathode and to avoid Pb precipitation in soil cells, 1 mol/L HNO_3 is added in the cathode reservoir so as to prevent the movement of the base front and increase the electric conductivity of the soil cell close to the cathode. Therefore, the production of OH^- ions at the cathode by the water electrolysis is theoretically evaluated using the Faraday equation. With a 0.6 A current, the estimated rate of OH^- is 6.2×10^{-6} mol/s. The corresponding flow of 1 mol/L HNO_3 needed at the cathode to neutralize OH^- ions is 0.02 L/h as determined by Eqn. (1):

$$n = It/F \quad (1)$$

where n is the mole number of OH^- ions; I is current, A; t is duration of treatment, s; F is Faraday constant.

Fig. 7 shows the variation of the soil cell pH value versus time and distance from the anode. Overall, the whole soil pH values are decreasing during the treatment process. After 30 h of treatment, the soil pH value near the anode (2 cm) is reduced to less than 1 because of the H^+ production via the electrolysis reaction. This acid front moves toward the cathode. The cathode reservoir pH value decreases gradually from 6.96 to 1.90 because the OH^- production is strongly buffered by HNO_3 added to it. The final soil pH value is between 1.2 and 1.9.

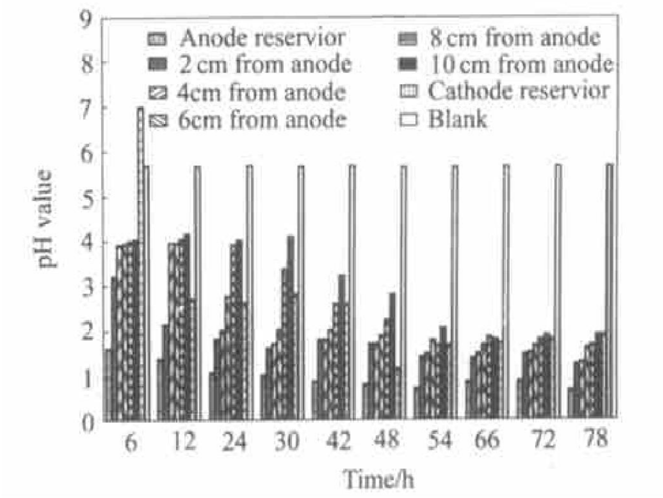

Fig. 7 pH value of soil cell versus time

Fig. 8 shows the evolution of tank voltage versus time. On constant current condition in the experiment, the tank voltage decreases sharply in 0 − 6 h from 117.0 V to 22.7 V, then the decrease slows down in the following process. This result shows that controlling pH value of the cathode can efficiently improve the electric conductivity of the soil cell.

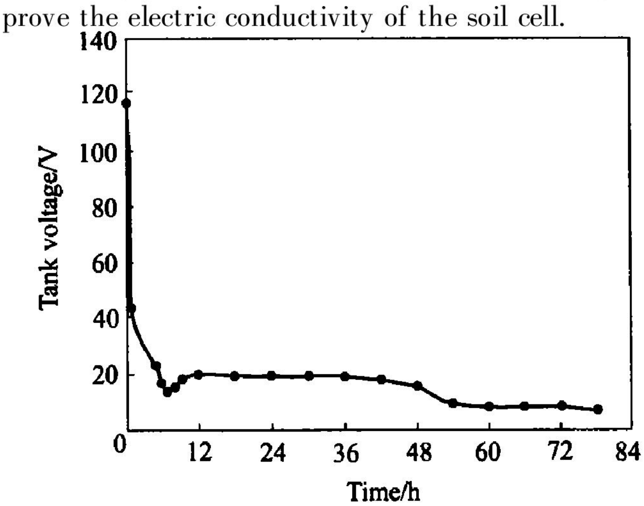

Fig. 8 Tank voltage versus treatment time

After 72 h of treatment, Pb concentrations decreased obviously as shown in Fig. 9, and the average Pb removal efficiency is 79.5%. The Pb concentrations of each soil cell are not the same at the beginning of the experiment. It is due to the free diffusion of Pb^{2+} at the water-saturated condition.

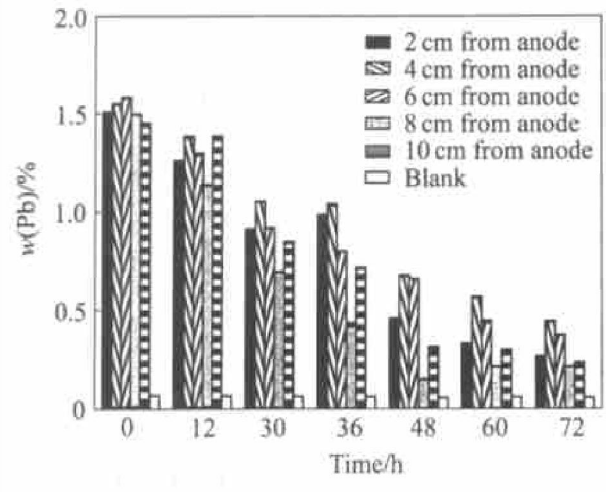

Fig. 9 Pb concentration of soil cell versus time

With a constant current of 0.6 A, an average voltage of 15.2 V, and treatment duration of 72 h, the calculated energy consumption is 0.6 kW·h. Since this amount of energy is used to treat 0.002 1 m^3 of the red soil, the energy consumption is 285 kW·h per m^3 red soil.

4 CONCLUSIONS

1) The electrokinetics technology is suitable for remedying Pb contaminated red soils.

2) Cathode controlling with HNO_3 is effective for improving removal effects. It can avoid Pb precipitating at the soil cells close to the cathode region, increase removal efficiency, decrease tank voltage and save the energy consumption.

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