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Research paper

The influence of macroelements on energy consumption during periodic power electrokinetic remediation of heavy metals contaminated black soil

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ABSTRACT

Two periodic power (12 h ON/12 h OFF and 18 h ON/6 h OFF) electrokinetic (EK) treatments were conducted to investigate their influence on the removal of heavy metals (Cd, Cu, Ni, Pb and Zn), depletion of soil macroelements (K, Na, Ca, Mg, Fe and Mn) and electric energy consumption. The results indicated that the removal efficiencies of heavy metals were higher than 80% in all the tested EK treatments. Periodic power EK treatment reduced the depletion of Fe and the electrical energy consumption compared with the constant power EK treatment. The lowest energy consumption and depletion of Fe were obtained in the periodic power of 12 h ON/12 h OFF EK treatment. A specific energy concept was developed based on the assumption that all the metals were removed by electromigration and used to predict the energy consumption for each metal. The results showed that more than 40% of electric energy was consumed to remove Fe in all the tested EK treatments. In comparison with the constant power EK treatment, the energy consumption for Fe reduced by 1264 kWh/m³ in the periodic power (12 h ON/12 h OFF) EK treatment, which accounted for 54.74% of the total reduction energy consumption in this EK treatment. Our results demonstrated that the reduction of energy consumption for Fe was one of the main contributors to save the total energy consumption.

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1. Introduction

Electrokinetic (EK) remediation is a promising remediation technology and widely used for extracting heavy metals from soils [1,2]. It applies an electrical potential gradient to carry and remove heavy metals through electromigration and electroosmotic flow (EOF) [3]. *In-situ* and *ex-situ* EK remediations have been reported for removal of heavy metals contaminated soils [4,5]. In the previous studies, most researchers focus on the removal of the target heavy metals during the EK remediation process. However, EK remediation may deplete the macroelements in soil, which should be avoided as most of them are essential elements for plant growth [6,7].

High cost of EK remediation becomes a major barrier to its widespread application [8]. The total cost of the EK remediation mainly includes fabrication and installation of electrodes, electric energy, labor and other fixed cost, et al. [9,10]. Among these

economic factors, electric energy consumption is a dominant factor that affects the total cost of the EK remediation [9]. Many factors including soil characteristics, contamination conditions, and electrode configuration or spacing significantly influence the energy consumption during the EK remediation process [11]. The soils with high ionic strength increased the electrical conductivity, thus more electric energy might be wasted in dissolving and carrying macroelements out of the soil [9,11]. In the previous study, electric power consumption per cubic meter of treated soil and per unit mass of heavy metals are used to calculate the total electrical energy consumption [12]. However, these methods are hard to compare the electrical energy consumption for each heavy metal or macroelement in the multi-heavy metals contaminated soil. Thus, it is necessary to develop a method to predict the electrical energy consumption for the removal of each metal. It is helpful to understand the influence of removal of each metal on the total energy consumption.

To reduce the energy consumption of EK remediation, pulsed electric field has been applied to EK systems [13,14]. Pulsed electric field has significant advantage in the EK remediation of heavy metals and saline from soil. The application of pulsed power “OFF

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time” can diminish the negative influences of the polarization effect (produced during the period with current “ON”), such as activation, resistance, and concentration polarization [10,15–17]. Many researchers reported that pulsed EK remediation have similar heavy metals removal efficiency compared to constant EK remediation but provide noticeable electrical energy savings under identical conditions [18,19]. Additionally, the pulsed electric field could control slight changes of pH and electric conductivity and effectively prevent electrode corrosion compared with the constant EK process [19]. Meanwhile, it has been proved that the pulsed EK process decreased the volume of EOF [20,21]. However, it is still unknown whether the pulsed EK remediation can reduce the removal of macroelements from the soil.

Thus, the periodic power EK remediation was carried out to investigate the influence on removal of heavy metals (Cd, Cu, Ni, Pb and Zn), depletion of the macroelements (K, Na, Ca, Mg, Fe and Mn) and energy consumption. The constant power EK remediation was conducted for comparison. Meanwhile, a simple model for the prediction of the energy consumption for each metal was developed and tested by comparing the energy consumption in this study and our previous study [22]. Then, the model was applied to predict the energy consumption for each metal in different EK treatments. We aim to demonstrate the relationship between removal of the metals and energy consumption in the periodic power EK remediation.

2. Material and methods

2.1. Chemicals

All the chemicals used in the current study were analytical grade. HCl was purchased from Beijing Chemical Works (China). All the other chemicals were obtained from Sinopharm Chemical Reagent Co., Ltd (China). Standard solutions of heavy metals and macroelements were purchased from the National RM Resources Network. All the solutions were prepared used Milli-Q water.

2.2. The preparation of heavy metal-contaminated soil

Black soil was collected from arable surface soil (10–20 cm depth) in Northeast Institute of Geography and Agroecology in Jilin province. The soil sample was air-dried and grounded to pass through a 0.84 mm sieve [23]. The main physical and chemical properties of collected natural soil were analyzed and summarized in Table 1. Heavy metals contaminated soil was obtained by spiking

the nitrates of Cd(II), Cu(II), Ni(II), Pb(II) and Zn(II). In brief, nitrate salts of these heavy metals were dissolved in Milli-Q water. The collected soil and the prepared solution were mixed thoroughly with the rate of soil: solution (m/V) = 1:1 and maintained in a pot at room temperature for 1 month. Thereafter, the heavy metals contaminated soil was air-dried again and sieved through a 0.84 mm sieve. The total concentrations of metals and their fractionations of contaminated soil were analyzed. A total of 320 g of contaminated black soil was loaded into the test cell using a tamper.

2.3. EK experiments

The EK cell was constructed using polymethyl methacrylate which was divided into three compartments: a soil cell and two electrode chamber (Fig. S1) [24]. The soil cell (15 cm × 3.8 cm × 6 cm) connected to the electrode compartments in both ends, using perforated polymethyl methacrylate and a filter paper. All the experiments were conducted at room temperature, without pH control. Graphite electrodes (7 cm × 3.5 cm × 1 cm) were used for the anode and cathode, respectively. The solution of 0.2 mol/L citric acid (CA) mixed with 0.05 mol/L CaCl₂ was used as anolyte and catholyte throughout the experiment [22]. Each electrolyte reservoir was filled with 600 mL of anolyte or catholyte and was replaced with fresh solution every five days. The electrolytes were circulated from electrolyte reservoir into the corresponding electrode chamber using peristaltic pump at the rate of 10 mL/min and the electrolyte level in the electrode chamber was maintained equally. A constant power (EK1), periodic 18 h ON/6 h OFF (EK2) and periodic 12 h ON/12 h OFF (EK3) power with a steady potential gradient of 1 V/cm were applied for 705 h. During the experimental period, electric current was monitored by using a logger (MW100, Yokogawa Electric Corporation, Japan). When EK experiments were terminated, the soil sample in each EK system was sectioned into five equal parts and labeled as S1 to S5 from anode to cathode.

2.4. Analytical method

The soil pH was measured using a pH meter (S220 seven compact, Mettler-Toledo International Inc., Switzerland) by preparing slurries with soil to water ratio of 1:2.5 [25]. Water-soluble anions (Cl⁻, SO₄²⁻, NO₃⁻) were analyzed using ion chromatography (ICS-900, Dionex, USA). The total metals concentrations in soil were determined by digestion with HNO₃ + HClO₄ + HF [26]. Metals speciation in the soil were determined through a three-step sequential extraction according to the procedure recommended by the Standards, Measurements and Testing (former BCR) Programme of the European Commission. In this approach, metals were divided into exchangeable fractions (F1), reducible fractions (F2), oxidisable fractions (F3) and residual fractions (F4) [27]. The detailed sequential extraction procedure was described in Table S1 [28]. The concentrations of metals ions in the supernatant were filtered through a 0.22 μm filters and analyzed by an Inductively Coupled Plasma Optical Emission Spectrometer (ICP-OES, ICP-5000, Focused Photonics, Inc., China).

2.5. Power consumption

In the previous study, most of researches just calculate the total electrical energy consumption in the EK remediation [24]. For the multi-heavy metals contaminated soil, it is hard to compare the electrical energy are consumed to remove each heavy metal [22]. Meanwhile, it is unclear that how much electrical energy is consumed to remove macroelements. Thus, in this study, we

Table 1
The properties of the natural black soil.

Characteristics	Value
Particle size analysis (%)	
Sand (%)	21.83
Silt (%)	64.68
Clay (%)	13.49
Organic matter (%)	2.416
CEC (cmol/kg)	15.72
pH	6.78
Electrical conductivity (μS/cm)	3.8
Moisture Content (%)	17.1
Porosity (%)	54.67
Hydraulic conductivity (cm/s)	0.0014
Metals total concentration (mg/kg)	
Cd	0.049
Cu	20.32
Ni	29.24
Pb	17.55
Zn	52.46

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