



Can electrokinetic removal of metals from contaminated paddy soils be powered by microbial fuel cells?



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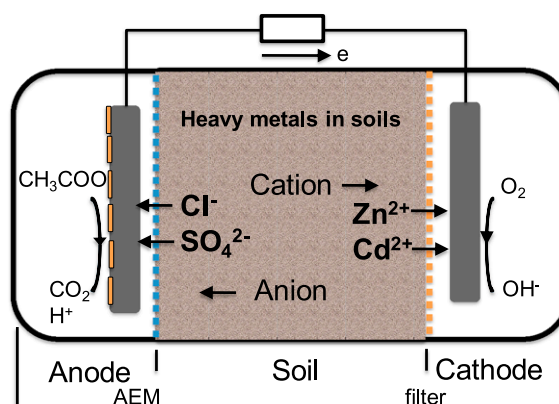
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HIGHLIGHTS

- Microbial fuel cells were applied for soil electrokinetic remediation.
- Zinc and cadmium were significantly removed from contaminated soils.
- A low cost and self-powered electrokinetic remediation system is reported.

GRAPHICAL ABSTRACT



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ABSTRACT

Soil electrokinetic remediation (ER) is an efficient technique to remove heavy metals from the complex soil matrix, however, this process needs a large amount of electricity input, which increases the cost and limits its application in remote agricultural fields. Microbial fuel cells (MFCs) are able to convert organic matter into electric current by using extracellular-respiring microbes. In this study, MFCs were designed and applied to electrokinetic removal of zinc (Zn) and cadmium (Cd) from a contaminated paddy soil. The current produced by three-chamber MFCs was $\sim 0.4 \text{ A m}^{-2}$ when loaded with the citric acid-amended soil. Replacement of the anion exchangeable membrane with nylon filter increased the transport rate of metal ions from the middle remediation chamber to the cathode chamber. After 78-day running, the systems with 300Ω resistors removed 12 mg Zn and 0.7 mg Cd more than those without resistors, suggesting that the current produced by the MFCs could significantly facilitate metal removal. The BCR extraction methods were applied to characterize the metal fractionation after ER treatment. The results showed that most of the removed metals came from acid extractable and reducible fractions. This study provides evidences that MFCs could power the soil ER processes, but the removal efficiency needs to be improved before its application in contaminated fields.

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1. Introduction

Novel technologies for the remediation of soils contaminated with heavy metals (HMs) are highly required due to increasing concern of HMs in agricultural soils. Concentrations of HMs in soils are elevated via various pathways, such as atmospheric deposition, livestock manures, fertilizers and agrochemicals, and sewage irrigation (Luo et al., 2009). Soil contamination with HMs can reduce crop yields and food quality, and eventually increase health risks to humans through food chain (Cui et al., 2004).

Soil or sediment is complex and often serves as a sink of metals. When metal ions are introduced into soil system, they exist as free ions, or bind with minerals and organic matter, or integrate into the mineral crystals (Bradl, 2004). The bioavailability and mobility of metals could be partly estimated by their fractions in soils (Ahumada et al., 2011). The complex fractionation of metals in soils made it difficult for their removal from the complex soil system. Electrokinetic remediation (ER) is a promising technology for contaminated site. This technique uses electric current to drive the movement of charged ion and remove the heavy metal ions from the low porosity soils (Lu et al., 2012). The removal efficiency of HMs was normally more than 70% of the total HMs (Virukutyte et al., 2002). However, electrokinetic remediation costs a lot of electric energy, varying from 38 to 2760 kWh/m³ for urban contaminated sites (Cang and Zhou, 2011). Thus, it is not suitable for large scale operation at remote area, e.g. remediation of agricultural field.

Self-powered technologies is an alternative method assisting in situ soil ER process. Renewable energy sources like solar cells are good candidates for self-powered ER (Baek et al., 2012). The performance of solar cells-assisted ER process is comparable with DC power supply (Yuan et al., 2009). However, the dependence of light intensity limits the application of solar cells in soil ER. Self-powered ER also can be achieved by using galvanic cells. A tentative study by imbedding iron in sediment as a sacrificial anode for removing copper was carried out (Yuan et al., 2009). The combination of self-powered technologies and soil ER are still under development.

Organic matter (OM) in agricultural area is abundant and can be converted into electricity by microbial fuel cells (MFCs) (Chen et al., 2012). A common MFC is constituted of an anode chamber and a cathode chamber. In the anode chamber, the extracellular respiring bacteria grew with OM supply and oxidize it into CO₂. The electrons derived from OM move through external circuit to the cathode chamber and are accepted by the oxidizing chemicals, such as oxygen, ferric iron and nitrate. The electricity produced by MFCs has been successfully used to power the small-scale device in remote areas (Tender et al., 2002), stimulate the degradation of organic pollutants (Wang et al., 2012), and remove salt from saline water (Cao et al., 2009). However, the efficiency of the MFC producing electricity for electrokinetic removal of heavy metals from soils is unclear.

In this study, we applied MFC as the only power source for electrokinetic remediation of the field collected paddy soil contaminated with high levels of heavy metals (Zn and Cd). The Zn and Cd transportation between bulk soil and cathode chambers was monitored during the experiment. The remediation efficiency was evaluated by measuring the HMs transported to cathode chamber and the metal fractionations before and after ER treatment with MFCs.

2. Materials and methods

2.1. Microbial fuel cell construction

The three chamber MFCs are constructed as described by Cao et al. (2009). The anode and cathode chambers were hollow cubes with a hole of 30 mm in length and 30 mm in diameter. The middle remediation chamber was a cylinder of 10 mm in length and 30 mm in diameter. An anion exchange membrane (AEM, AMI-7001S, Membranes International Inc.) and a cation exchange membrane (CEM, CMI-7000S, Membranes International Inc.) were used to separate the three chambers. A graphite mat (rectangle, 3 mm × 30 mm × 90 mm, Beijing Sanye Carbon Co., Ltd, Beijing, China) was rolled into the anode chamber serving as anode. The cathode was a graphite mesh (Xiamen Fusheng Composite Co. Ltd) coated with Pt catalyst (0.3 mg/cm²). During the start stage, 10 g l⁻¹ NaCl solution was periodically added in the remediation chamber as supporting electrolyte. The salinity was measured by using an electric conductivity meter (sensION 156, HACH Co., CO, USA). The external resistances were 300 Ω. Cell voltages were recorded by using a data acquisition board (NI USB6225, National Instruments Corporation, USA).

The supernatant from paddy soil was inoculated into the anode chamber. The anode medium contained 2 g sodium acetate, 4.4 g KH₂PO₄, 3.4 g K₂HPO₄ · 3H₂O, 1.5 g NH₄Cl, 0.1 g MgCl₂ · 6H₂O, 0.1 g CaCl₂ · 2H₂O, and trace mineral nutrients in 1 l deionized water, which were described previously (Cao et al., 2009). The anode solution was replaced when the cell voltages dropped to around 50 mV. The cathode chamber was connected to a bottle of 500 ml cathode medium (1 mM KH₂PO₄) with continuing aeration. The cathode medium was cycled between the cathode chamber and the 500 ml store bottle by using a peristaltic pump. The pH of cathode medium was measured daily and adjusted to 3 with 20% nitric acid if necessary.

2.2. Soil remediation

A paddy soil with metal contamination was collected from Xinma village, Hunan province. The soil is categorized into Fec Stagnic Anthrosols (Chinese Soil Taxonomy) or Hydragric Anthrosols (World Reference Base for Soil Resources). The soil sample was air-dried and passed through a 2-mm sieve before use in experiments. Zn and Cd concentrations in the soil were 11.6 and 0.84 g/kg, respectively. Fifty-six gram dry soil was mixed with 88 g 100 mM citric acid solution to make the water-saturated sludge. The citric acid was chosen because it was environmentally friendly for soil remediation and can release Zn and Cd ions from solid soil particles (Giannis and Gidarakos, 2005). Approximately 12 g sludge was squeezed into the middle remediation chamber of each cell. The remaining sludge was stored in a sealed centrifuging tube as control. To maintain the soil humidity, a syringe with 1 ml 100 mM citric acid was connected with the middle chamber through the upper hole. After filled with the contaminated soils, two cells were kept running with 300 Ω resistance (close treatment), and another two runs without external resistance (open treatment). The concentrations of Zn and Cd in anode and cathode chambers were analyzed every 3 days by using inductively coupled plasma-mass spectroscopy (ICP-MS, Agilent 7500, Agilent Inc.). The cathode solution was replaced every 2 weeks.

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