

# Remediation of soil co-contaminated with petroleum and heavy metals by the integration of electrokinetics and biostimulation



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## HIGHLIGHTS

- Regular electrolyte refreshment is favorable for maintaining high electric current.
- EDTA-injection could greatly reduce Pb ecotoxicity.
- Although adding Tween 80 reduced current, it could enhance microbial growth and biodegradation.
- Oil biodegradation degree increased toward the anode and cathode.
- Microbial action was beneficial to metal release from soil matrix.

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## ABSTRACT

Successful remediation of soil co-contaminated with high levels of organics and heavy metals is a challenging task, because that metal pollutants in soil can partially or completely suppress normal heterotrophic microbial activity and thus hamper biodegradation of organics. In this study, the benefits of integrating electrokinetic (EK) remediation with biodegradation for decontaminating soil co-contaminated with crude oil and Pb were evaluated in laboratory-scale experiments lasting for 30 days. The treated soil contained 12,500 mg/kg of total petroleum hydrocarbons (TPH) and 450 mg/kg Pb. The amendments of EDTA and Tween 80, together with a regular refreshing of electrolyte showed the best performance to remediate this contaminated soil. An important function of EDTA-enhanced EK treatment was to eliminate heavy metal toxicity from the soil, thus activating microbial degradation of oil. Although Tween 80 reduced current, it could serve as a second substrate for enhancing microbial growth and biodegradation. It was found that oil biodegradation degree and microbial numbers increased toward the anode and cathode. Microbial metabolism was found to be beneficial to metal release from the soil matrix. Under the optimum conditions, the soil Pb and TPH removal percentages after 30 days of running reached 81.7% and 88.3%, respectively. After treatment, both the residual soil Pb and TPH concentrations met the requirement of the Chinese soil environmental quality standards.

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## 1. Introduction

Soil contamination by heavy metals and organic pollutants has become major environmental and human health concerns worldwide, due to rapid industrialization and urbanization [1]. The remediation of soils co-contaminated by organics (petroleum, chlorinated solvents, pesticides, herbicides) and heavy metals is a complex problem, as the chemical processes and remediation technologies are different for each group of pollutants [2].

Petroleum contamination has been a major environmental concern for decades due to increased dependence on petroleum products around the world [3,4]. Bioremediation is a popular and sustainable approach for restoring petroleum-contaminated soil because of low cost and preserving the quality and functions of soil [5]. Nevertheless, petroleum and heavy metals may co-exist in nature in some cases [6,7]. Some studies have demonstrated that high levels of heavy metals can suppress oil biodegradation in the environment due to the presence of toxic metals inhibiting the activity of the degrader bacteria [6,8].

Electrokinetic (EK) remediation is a green remediation technology developed in recent ten years, which can promote the degradation and/or the removal of organic and metal contaminants

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[9]. As compared to conventional remediation methods, EK remediation offers the advantages including simplicity, safety, wide ranges of contaminated media and contaminants, flexibility and cost-effectiveness [10]. Previous studies have shown that EK technology has the potential to remove heavy metals [11], organic pollutants [12–14], and their mixture [15–20] from the contaminated soils.

The integration of EK and biological techniques (also called bioelectrokinetics, BioEK) can make up for each other's limitations, and thus achieve the objective of enhancing remediation efficiency and lowering the energy costs. BioEK was first applied to the treatment of heavy metal-contaminated soils [9,21]. Nowadays, it is also used to remove organic pollutants from the contaminated soil, which including petroleum hydrocarbons, halogenated hydrocarbons, phenols and so on [22]. Nevertheless, little research has yet been performed on BioEK for the remediation of soil co-contaminated by heavy metals and petroleum.

The aim of the present laboratory study was to investigate the feasibility and efficiency of remediation of soil co-contaminated with crude oil and heavy metals simultaneously using the combination of EK and biological processes. Removal of metal was mainly through electromigration, while biodegradation of oil was realized by biostimulation of the indigenous microorganisms. The overall BioEK results were compared with those of abiotic EK and biological treatment alone.

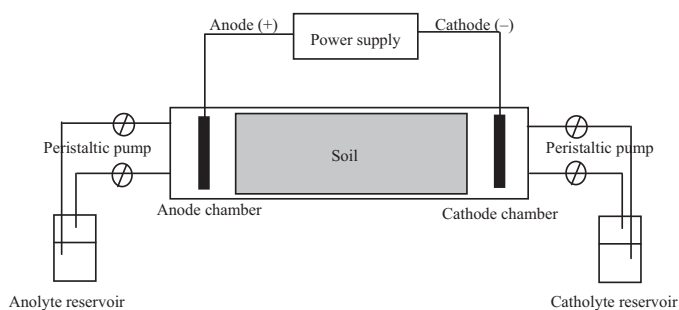
## 2. Materials and methods

### 2.1. Co-contaminated soil

The soil used in this study was collected from a petroleum refinery plant located at Tianjing, China. The soil had been contaminated over ten years with crude oil and heavy metals from leakage, spillage and maintenance activities. Soil samples were collected in plastic bags, thoroughly mixed, sieved through 2 mm pore size mesh, and stored at 4 °C. Table 1 lists the main physical, chemical and microbiological characteristics of the contaminated soil. Concentrations of Pb and hydrocarbons exceeded Chinese action levels [23]. The basic soil properties were measured according to standard methods [24].

**Table 1**  
Characteristics of the soil used for the experiments.

Characteristics	Value
pH (1:2 soil:water)	6.7
Conductivity ( $\mu\text{S}/\text{cm}$ )	1130
Water holding capacity (wt%)	41.2
Humidity (wt%)	17.5
Permeability coefficient (m/S)	$2.1 \times 10^{-7}$
Sand (wt%)	58.3
Silt (wt%)	26.4
Clay (wt%)	15.3
Texture	Loamy sand
Total organic carbon (wt%)	8.6
Total N (mg/kg)	65
Total P (mg/kg)	14
K (mg/kg)	38
Ca (mg/kg)	115
Mg (mg/kg)	12
Cu (mg/kg)	6
Pb (mg/kg)	450
Heterotrophs (CFU/g)	$5.5 \times 10^6$
Hydrocarbon degraders (CFU/g)	$3.6 \times 10^4$
TPH (mg/kg)	12,500
Saturated hydrocarbons (mg/kg)	9130
Aromatic hydrocarbons (mg/kg)	3370
Resin-asphaltene (mg/kg)	1420



**Fig. 1.** A schematic diagram of the lab-scale electrokinetic reactor.

### 2.2. Electrokinetic apparatus

The schematic diagram of the lab-scale EK reactor is shown in Fig. 1. The EK reactor consists of five compartments: the anolyte reservoir (1 L), anode chamber (4 cm × 4 cm × 4 cm), soil cell (20 cm × 4 cm × 4 cm), cathode chamber (4 cm × 4 cm × 4 cm) and catholyte reservoir (1 L). A Pt-coated titanium and carbon plate were used for the anode and cathode, respectively. Filter paper was placed between the soil and electrode compartments to prevent soil particles from penetrating into the electrolyte solution reservoirs.

### 2.3. Experimental design

Before being packed in the cell, the soil was saturated with inorganic salts solution, which contained (g/L):  $\text{KH}_2\text{PO}_4$  0.9,  $\text{K}_2\text{HPO}_4$  0.1,  $\text{NH}_4\text{NO}_3$  0.1,  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$  0.1,  $\text{CaCl}_2$  0.080,  $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$  0.01 [25].

A series of experiments were performed by varying treatment conditions. Each soil cell was loaded with 600 g of the soil. One contaminated soil microcosm amended with  $\text{NaN}_3$  (1.0 wt.%) was used for abiotic process measurements (abiotic control). In addition, to estimate the pollutant losses mainly through microbial action, another control microcosm was set up for the contaminated soil (biotic control). The biotic-control soil was supplemented with a certain amount of  $\text{NH}_4\text{NO}_3$  and  $\text{K}_2\text{HPO}_4$  to give a final C:N:P ratio of 100:10:1 [26]. The above two control microcosms received no application of electric current and electrolyte, and soil moisture content was kept at approximately 20% (w/w) by regular addition of sterile de-ionized water.

Electrolyte was prepared to supply the bacterial consortium with ionic nutrients, to increase the electrical conductivity of soil, and to control pH. The composition of electrolyte is as follows (g/L):  $\text{K}_2\text{HPO}_4$  87.0,  $\text{KH}_2\text{PO}_4$  68.0,  $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$  0.2,  $\text{NH}_4\text{NO}_3$  1.0, pH 8.0 (anode) or 5.6 (cathode). The anolyte and catholyte reservoirs were filled with the electrolyte solutions, which were recirculated independently by the peristaltic pumps. To improve metal removal, disodium salt EDTA ( $\text{C}_{10}\text{H}_{12}\text{N}_2\text{Na}_2\text{O}_8 \cdot 2\text{H}_2\text{O}$ ) used as the conditioning agent was added in the catholyte reservoir (BioEK2 test) to reach a concentration of 0.1 M. To enhance oil removal, Tween 80 was used as the anolyte conditioning agent at a concentration of 2.0 g/L (BioEK3 test). After the addition of conditioning agent, the pH of electrolytes was adjusted to 8.0 (anode) or 5.6 (cathode). The electrolyte solutions were refreshed every three days to maintain consistent properties. Additionally, two contaminated soil microcosm amended with  $\text{NaN}_3$  (1.0 wt.%) was loaded into the EK reactor to exclude potential biotic interferences on the metal removal without (EK1) or with electrolyte refreshment (EK2). All soil treatments were carried out at room temperature ( $24 \pm 2$  °C) and run in triplicate, which are summarized in Table 2.

A constant potential difference of 40 V (2.0 V/cm) was applied to all electrokinetic experiments. Electric current was measured once a day using a current meter. Electroosmotic flow (EOF) was calculated from the difference in volume of the catholyte reservoir.

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