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Research article

## Simultaneous removal of polycyclic aromatic hydrocarbon and heavy metals from an artificial clayey soil by enhanced electrokinetic method



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#### ABSTRACT

Contamination of soil by a mixture of organic and non-organic pollutants due to various anthropogenic and natural causes is one of the most important issues in soil pollution. Generating a single layer of soil is a time-consuming process, and soil is a significant part of the environmental cycle; thus, protection and remediation of soil are of paramount importance. In this study, phenanthrene (PHE) as a representative for polycyclic aromatic hydrocarbon and lead (Pb), zinc (Zn), and nickel (Ni) as heavy metal pollutants were used because of their synergistic effects and abundant presence in soil. Soils with three different organic matter levels (OMO, OM1, and OM2) were prepared, spiked with pollutants, placed in electro-kinetic cells, and remediated for 15 days. To improve the remediation process and increase the efficacy of pollutant removal, pH control methods and extracting agents were used. Two non-ionic surfactants, Tween 80 and Brij 35, and one chelating agent, EDTA, were utilized for this purpose. A total of nine tests were performed. The soil was divided into five equal sections at the end of each test, and the pH and concentration of pollutants were analyzed. Results indicated that the highest removal percentage of PHE was accomplished by Brij 35 (61%) in OM0. Increasing organic matter resulted in decreasing PHE removal. In this regard, Tween 80 showed enhanced performance. The order of removal of heavy metals was zinc > nickel > lead, and zinc had the highest removal rate of 61%.

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### 1. Introduction

Organic and non-organic contaminants such as polycyclic aromatic hydrocarbons (PAHs) and heavy metals are some of the pollutants that have raised concern because of their toxicity and nature, which pose danger to the environment and humans. In addition to natural sources, anthropogenic sources include industrial, mining, and agricultural practices; aerosols from fuel combustion; and pipeline transportation of oil. These sources contribute to environmental pollution, specifically to soil. The coexistence of these two pollutants in contaminated soil, such as Manufactured Gas Plant (MGP) sites, is prevalent (Alcántara et al., 2012; Nadal et al., 2011; Reddy et al., 2006; Saison et al., 2004; Thavamani et al., 2012). The simultaneous presence of these two contaminants in soil, which exhibit different characteristics but synergistic effects, makes conventional remediation a difficult task in low-permeability soil (Alcántara et al., 2012; Maturi et al., 2009). Therefore, new remediation techniques have been developed to address this issue, and electrokinetic remediation has shown promising results and is still in the development stage (Kuppusamy et al., 2017).

Electrokinetic remediation is a technology that has demonstrated satisfactory results in removing a single pollutant from soil and is applicable to low-permeability soils and a mixture of contaminants (Lopez-Vizcaino et al., 2014; Saichek and Reddy, 2004; Yuan and Weng, 2004). Its principle is based on generating an electric field by applying a weak direct electric current via two opposite electrodes and inducing the transportation of pollutants to the electrodes through different mechanisms such as

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electroosmosis, electromigration, electrophoresis, and advection (Acar and Alshawabkeh, 1993; Falcigila et al., 2017; Virkutytea et al., 2002). Several phenomena such as sorption/desorption, precipitation, and dissolution are responsible for the migration of contaminants (Puppala et al., 1997; Yuan and Weng, 2006). One of the most important and influential reactions in electrokinetic remediation is water electrolysis (Acar and Alshawabkeh, 1993; Amrate et al., 2005: Virkutytea et al., 2002). As a result of this reaction, hydrogen and hydroxyl ions are generated in anode and cathode chambers, respectively, and they migrate to the opposite electrodes and change soil pH. The physicochemical changes in unenhanced electrochemical remediation cause constraints in soil decontamination. To overcome limitations generated by some of these phenomena, enhanced techniques are necessary. These techniques are categorized into three groups: solubilizing contaminants and keeping them in the liquid phase, controlling soil pH, and coupling other remediation methods with electrochemical remediation (Yeung and Gu, 2011).

Previous studies have shown that electrochemical remediation has the potential to remove heavy metals and organic contaminants from soil when they are separate in soil. However, the conditions differ in their simultaneous existence. To date, few researchers have focused on the simultaneous remediation of mixtures of PAH and heavy metals by electrokinetic remediation (Cang et al., 2013; Zou et al., 2016).

This study aimed to investigate the applicability of different enhancement agents and pH control on the simultaneous removal of a mixture of PAH and heavy metals from artificial clayey soil. To avoid complications associated with natural soil, a real soil situation was reconstructed. Pollutants were mixed, and different percentages of sand and clay were used. Low permeability was ensured. Researchers illustrated that soil organic matter plays a crucial role in remediation and removal of contaminants from soil (Medina et al., 2018). Few studies have been carried out to investigate the effect of organic matter on removing heavy metals and PAHs, so this work aimed to fill this gap. To the best of the authors' knowledge, this study is the first to consider the effect of organic matters on the simultaneous removal of heavy metals and PAHs.

#### 2. Materials and methods

#### 2.1. Artificial soil

Kaolin was selected for the tests because of its low permeability and low buffering capacity. Artificial soil samples were provided from the Indian International Laboratory. X-ray diffraction and Xray fluorescence spectroscopy analyses were carried out to determine the mineralogical and chemical compositions of the artificial soils. The used kaolin was 100% kaolinite, and the sand was a mixture of quartz, calcite, and dolomite. The results are presented in Table 1. A laser diffraction particle size analyzer (LA-950, Horiba) was used for soil grading. Approximately 100% of kaolin particles were smaller than 20  $\mu$ m, nearly 60% were smaller than 2  $\mu$ m, and 100% of sand particles were smaller than 2 mm. Fig. 1a and b show the kaolin and sand particle size diagrams, respectively.

#### 2.2. Experimental apparatus

A photograph and schematic of the experimental electrokinetic setup are shown in Fig. 2. The rectangular cell was made of Plexiglas and was 28 cm in length, 7 cm in width, and 7 cm in height. The dimensions of the soil compartment and electrolyte solution reservoirs were  $20 \text{ cm} \times 7 \text{ cm} \times 5 \text{ cm}$  and  $4 \text{ cm} \times 7 \text{ cm} \times 7 \text{ cm}$ , respectively. A constant voltage gradient of 1.5 v/cm was generated by a D.C. power supply (0–40 V, 0–5 A). Both electrodes in anode

#### Table 1

Mineralogical and chemical composition of artificial soil samples.

Analyses		Kaolin	Sand
XRD analysis		Kaolinite	Major Phase: Quartz Minor Phase: Calcite Trace: Dolomite
XRF analysis	SiO2 (%)	44.39	88.36
	Al2O3 (%)	39.14	1.55
	Fe2O3 (%)	0.36	1.78
	CaO (%)	0.03	3.67
	Na2O (%)	0.02	0.02
	K2O (%)	0.01	0.13
	MgO (%)	0.01	0.28
	TiO2 (%)	0.386	0.168
	MnO (%)	0.005	0.019
	P2O5 (%)	0.066	0.029
	SO3 (%)	0.012	0.025

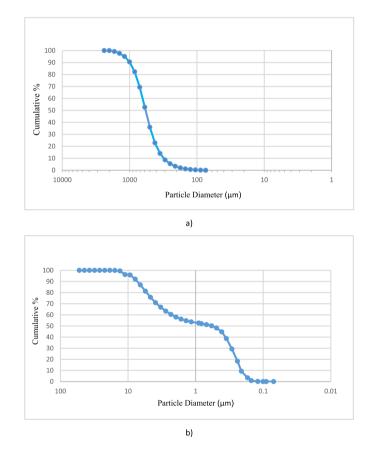


Fig. 1. Particle size diagrams of a) Sand, and b) Kaolin.

and cathode were reticular stainless steel (A316), and to facilitate pore fluid flow electrode container panels were perforated. Filter papers (Whatman no. 1) were used between electrodes and soil samples to minimize soil particles into the reservoirs. To circulate solution in reservoirs and avoid generating pH gradient, an air pump was used in the anode and cathode compartments.

#### 2.3. Artificial soil sample preparation

All artificial soil samples were composed of 60% kaolin and 40% sand, which were in the clayey soil category according to the USDA soil textural classification (USDA, 1987). To determine the effect of soil organic matter on the removal efficiencies, three artificial soil samples with different amounts of organic matter were prepared.

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