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# Electroremediation of a natural soil polluted with phenanthrene in a pilot plant

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#### HIGHLIGHTS

In pilot electroremediation plants, gravity and evaporation fluxes are even more relevant than electrokinetic fluxes.

- Desorption of PHE promoted by the electric heating seems to be a very significant removal mechanisms.
- PHE is also dragging with electroosmotic and electrophoresis fluxes in the electrolytic wells.
- After 2800 h of treatment, an average PHE removal of 25% is attained.

• To attain the complete remediation of the soil, the energy applied must be higher than 540 kWh m<sup>-3</sup>.

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#### ABSTRACT

In this work, a pilot plant with two rows of three electrodes in semipermeable electrolyte wells was used to study the electrokinetic treatment of a natural soil polluted with phenanthrene (PHE). The electrokinetic pilot plant was an open system, i.e., there was direct contact between the soil and air. To increase the solubility of phenanthrene, thereby enhancing its transport through the soil, an aqueous solution of the anionic surfactant dodecyl sulfate was used as a flushing fluid. The results show that at the pilot scale considered, gravity and evaporation fluxes are more relevant than electrokinetic fluxes. Contrary to observations at the laboratory scale, desorption of PHE promoted by electric heating appears to be a significant removal mechanism at the pilot scale. In addition, PHE is dragged by the electroosmotic flow in the cathodic wells and by electrophoresis after interaction of the surfactant with phenanthrene in the anodic wells. In spite of the long treatment time (corresponding to an energy consumption over 500 kWh m<sup>-3</sup>), the average removal attained was only 25%.

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#### 1. Introduction

Societal non-sustainable development has resulted in the release of a great variety of organic pollutants into soil and groundwater. The persistence of organic compounds in soil produces a complex impact that depends on numerous factors, including the physicochemical and biological properties of the pollutants, geological and chemical characteristics of the soil, the amount of soil contaminated and the quantity of pollutants. Some organic pollutants degrade in situ by indigenous microorganisms. The low solubility of other organics hinders their accessibility to microorganisms, preventing bio-degradation. A well-known group of this latter category of pollutants is the polycyclic aromatic hydrocarbons (PAHs), substances that are generally composed of non-polar molecules formed by two or more benzene rings [1]. PAHs are classified as hydrophobic organic compounds (HOC) that are highly carcinogenic, mutagenic and teratogenic [2,3]. Moreover, they are persistent in soils and sediments because of properties such as low volatility, low water solubility and low biodegradability. These characteristics inhibit their efficient removal from soil [4].

Recently, a wide variety of remediation technologies have been developed to address PAH soil pollution [5,6]. Among them, it is important to highlight the electrokinetic techniques, which are the result of different processes that occur in soil when a DC electric potential is applied through a group of electrodes located in the soil [7–10]. This flushing fluid-assisted technology has been particularly recommended for the treatment of low-permeability soil polluted with ionic and water-soluble compounds [11,12]. This







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technique can also be applied to the removal of hydrophobic compounds, although in this case, the non-polar character and slow desorption rates of the PAHs require the addition of special flushing fluids to increase the polarity of the pollutants, thus increasing their solubility in water. The special flushing fluids are usually solutions of surfactants and cosolvents [13], and they are able to improve the mass transfer of HOCs from the soil into the aqueous phase by decreasing the interfacial tension and by accumulating HOCs in the surfactant micelles [13–19]. Moreover, the addition of flushing fluids guarantees hydraulic saturation, enhances the conductivity of the soil and helps to maintain the ionic circuit in the soil.

In recent years, several works have focused on this topic. Most of them were performed at the laboratory scale [13–19], and typically, the soil electroremediation reactor consisted of a small hermetic column of less than 1 dm<sup>3</sup>, in which kaolin (or other clay or lime, not natural soil) was used as a model for low-permeability soil [13]. These studies have contributed to increase knowledge about the fundamentals of electrokinetic processes. However, the extrapolation of these results to an actual application without the appropriate scale-up would be risky, according to the results shown in the few works that assess the treatment of natural soil matrixes [2,20–22].

There is a plethora of factors that can influence the efficiency of the soil flushing electroremediation process, such as electrode material, electrode location, composition and flux of flushing fluids, electric field and operation time. Although the well-known iron sacrifice electrode is proposed in some applications, ideally, electrodes should be inert materials, such as platinum, silver, gold or graphite, to prevent the development of side reactions. Among these inert materials, and taking into account economic considerations, graphite is the most commonly used in practical applications [9], in spite of the well-known poor electrochemical stability for the large cell potentials applied in electrokinetic soil remediation processes  $(0.5-2.0 \text{ V cm}^{-1})$ .

With respect to the distribution of electrodes and the placement in the soil, there is great controversy. In some works, the use of electrodes in direct contact with the soil to promote the electroosmotic flux is proposed, although the electrical resistance can be high [23]. Another possibility is the insertion of the electrodes in electrolyte wells [24]. This latter option also facilitates the injection of flushing fluids. Although there are various possibilities, the use of wells containing a single cylindrical electrode appears to be the best choice. Regardless of the location of the electrodes, there are different electrode configurations, such as rows of facing electrodes and/or surrounding anodes with cathodes (or vice versa), with advantages in particular cases depending on the distribution of pollutants in the soil to be treated.

In this context, the goal of this work is to study the electrokinetic treatment of a natural soil polluted with phenanthrene (500 mg PHE kg<sup>-1</sup> soil). The electrodes were positioned into semipermeable electrolyte wells with a linear distribution (two rows of three facing electrodes). Both electrolyte wells and the soil were in direct contact with the atmosphere because the electrokinetic pilot plant was an open system. To increase the solubility of phenanthrene and thus to enhance its transport through the soil, aqueous solutions of the anionic surfactant dodecyl sulfate (10 g dm<sup>-3</sup>) were used as a flushing fluid.

#### 2. Materials and methods

#### 2.1. Materials

Soil from a quarry located in Toledo (Spain) was used in this study. This soil is characterized by its inertness, low hydraulic conductivity and lack of organic content. The mineralogical

Table 1

Mineralogica	l composition	and classification	by USCS.
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Mineral	%
Quartz	12
Feldspar	6
Calcite	1
Kaolinite	23
Glauconite	24
Muscovite	8
Montmorillonite	20
Smectite	-
Illite	6
Parameters	
Liquid Limit	42
Plastic Limit	24
Plasticity Index	18
USCS Code	CL
Low plasticity clay	

composition and the parameters used to classify this soil by the Unified Soil Classification System (USCS) are listed in Table 1.

Phenanthrene (97%) was selected as a model HOC. An anionic surfactant, sodium dodecyl sulfate (SDS), was used as the flushing fluid  $(10 \text{ gL}^{-1})$ . Their properties are listed in Table 2.

#### 2.2. Preparation of the polluted soil

The process of the preparation of the soil is important because of the complexity of natural soil. The process was divided into different stages: (1) positioning of three layers of gravel with different granularity for mechanical and drain support; (2) pollution of the soil with a phenanthrene/acetone solution and aeration of the spiked soil to promote evaporation of the acetone, as described in the literature [25–27]; (3) moistening the soil to 20% (a value fixed by the Proctor compaction study); (4) compaction of the soil in the electrokinetic reactor by compacting layers of a fixed height (5 cm) until achieving a density typical for natural soil (approximately  $1.4 \text{ g cm}^{-3}$ ).

#### 2.3. Experimental setup

The electrokinetic experiments were conducted in an electrokinetic remediation plant consisting of an electrokinetic reactor, a power source and tanks of electrolyte and surfactant. The reactor was a methacrylate prism with a soil capacity of  $175 \times 10^3$  cm<sup>3</sup> (LWH:  $70 \text{ cm} \times 50 \text{ cm} \times 50 \text{ cm}$ ). The electrodes used for both the anodes and the cathodes were graphite rods with dimensions  $1 \text{ cm} \times 10 \text{ cm}$ , positioned in semipermeable electrolyte wells, using an electrode configuration of rows facing the electrolyte wells, as described elsewhere [28,29]. The cathodic wells are connected to  $100 \text{ cm}^3$  sewers to accumulate the fluid transported through the soil and facilitate sampling. The reactor is designed to separate and collect the fluids through an outlet situated on the side wall of the reactor. Fig. 1 shows a schematic of the electrokinetic remediation plant.

Table 2		
Droportion	oftho	<b>C1</b>

Properties	ot	the	sur	fac	tanı

Surfactant	Sodium dodecyl sulfate
Туре	Anionic
Formula	C <sub>12</sub> H <sub>25</sub> NaO <sub>4</sub> S
MW	288.38
CMC/gl <sup>-1</sup>	2.3
Molecular structure	
0,0 0,0 0,0 0,0 0,0 0,0 0,0 0,0 0,0 0,0	

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