



## Feasibility of electrokinetic oxygen supply for soil bioremediation purposes



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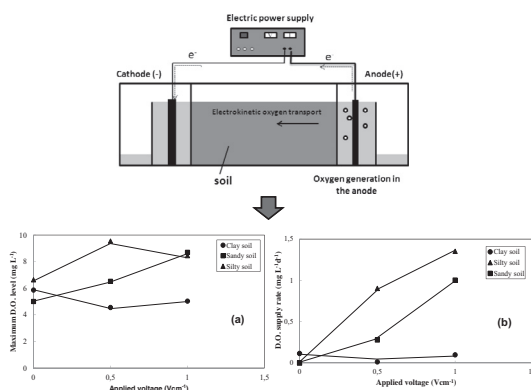
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### HIGHLIGHTS

- Feasibility of electrokinetic oxygen supply to soils was tested.
- Oxygen transport was available in silty and sandy soil but it was not in clay.
- Oxygen transport rate was low and depended on the voltage gradient.
- The method could be used for slow in situ biostimulation soil remediation processes.

### GRAPHICAL ABSTRACT



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### ABSTRACT

This paper studies the possibility of providing oxygen to a soil by an electrokinetic technique, so that the method could be used in future aerobic polluted soil bioremediation treatments. The oxygen was generated from the anodic reaction of water electrolysis and transported to the soil in a laboratory-scale electrokinetic cell. Two variables were tested: the soil texture and the voltage gradient. The technique was tested in two artificial soils (clay and sand) and later in a real silty soil, and three voltage gradients were used: 0.0 (control), 0.5, and 1.0 V cm<sup>-1</sup>. It was observed that these two variables strongly influenced the results. Oxygen transport into the soil was only available in the silty and sandy soils by oxygen diffusion, obtaining high dissolved oxygen concentrations, between 4 and 9 mg L<sup>-1</sup>, useful for possible aerobic biodegradation processes, while transport was not possible in fine-grained soils such as clay. Electro-osmotic flow did not contribute to the transport of oxygen, and an increase in voltage gradients produced higher oxygen transfer rates. However, only a minimum fraction of the electrolytically generated oxygen was efficiently used, and the maximum oxygen transport rate observed, approximately 1.4 mg O<sub>2</sub> L<sup>-1</sup> d<sup>-1</sup>, was rather low, so this technique could be only tested in slow *in-situ* biostimulation processes for organics removal from polluted soils.

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## 1. Introduction

Soil bioremediation consists of the elimination of pollutants present in polluted soils by microbial assimilation processes. These processes are based on the fact that the soil contains microbial consortia that are able to use the soil pollutants as a raw material and energy source for their growth and maintenance (Juwarkar et al., 2010). The principal advantage of soil biological treatment is its low cost, although it is slower than other physicochemical techniques, and it is always subject to the ability of the microorganisms to use the pollutants as a substrate. There are some basic requirements for successful biodegradation of the organic pollutants in soil: the presence of appropriate microorganisms, sources of energy, sources of carbon, nutrients, electron acceptors and suitable environmental conditions, such as temperature, pH and moisture (Bedient et al., 2000; Schmidt et al., 2007; Xu et al., 2010).

Bioremediation, as well as other soil remediation technologies, can be accomplished in two ways: *in-situ* and *ex-situ*. *In-situ* soil biological remediation does not require the removal of the polluted soil from its original site, which is an important advantage. However, this approach has additional challenges because the soil matrix is a complex and heterogeneous medium with different material and energy transport resistances. These resistances can hinder the interactions among the different agents that must take part in the degradation process. Because of these resistances, the input of nutrients to increase the rate of the degradation process can also be very difficult in medium- or low-permeability soils. This limitation is especially important in fine-grained soils such as clay.

In recent years, a new emerging *in-situ* hybrid technology has been proposed to increase the mobility and thereby the possibilities of interaction among microorganisms, pollutants and nutrients in the soil. This technique is called electrokinetic enhanced bioremediation or electrobioremediation (Wick et al., 2007). It is based on a combination of the electrokinetic mobility phenomena (electroosmosis, electromigration, and electrophoresis) that appear in a soil when a direct electric field is applied to it and the biological degradation process. In this technology, charged species can be moved throughout the soil matrix by electromigration and electrophoresis, while non-charged species can be dragged by the water mobilised by electroosmosis. Because of all these forms of transport, it is expected that interactions among the different agents involved in the degradation process could occur more frequently, increasing the rate of the degradation process (Luo et al., 2005; Chun et al., 2013).

Apart from the increase in the mobility of the species present in the soil matrix, electrokinetic processes can also be used to introduce nutrients into the soil, to ensure the correct balance between inorganic nutrients and organic pollutants, which would increase the rate and efficiency of biodegradation processes. Several authors have previously reported different studies investigating the supply of inorganic nutrients (nitrates, sulphates, phosphates, ammonium, etc.) by electrokinetic processes, showing promising results in some cases (Schmidt et al., 2007; Xu et al., 2010).

However, there are few studies about the influence of electrokinetic treatment on the dissolved oxygen (DO) concentrations of the groundwater of polluted soils (Franz et al., 2002). This is an important issue because biotransformations under aerobic conditions are more energetically favourable than the use of alternative electron acceptors, such as nitrate or sulphate (Spence et al., 2005). Due to the low diffusion rate of oxygen, it is a challenge to develop an appropriate alternative to supply a high enough DO concentration to meet the demand for *in-situ* soil aerobic remediation processes.

Different alternatives have been used to increase the concentration of DO in the media, such as bioventing, air sparging/biosparging or liquid delivery systems (Pardieck et al., 1992; Balcke et al.,

2004; Vogt et al., 2004). Additionally, several products, such as oxygen releasing compounds (ORC) and oxygen micro-bubbles have also been developed to oxygenate groundwater and/or soil (Kunukcu, 2007; Jechalke et al., 2010; Zawierucha and Malina, 2011; Chun et al., 2013).

In aqueous solutions, after applying an electric current, in addition to electrokinetic processes, water electrolysis takes place, generating hydrogen and oxygen at the electrodes according to the following equations:



Hydrogen is available at the cathode as an electron donor for reductive degradation processes, whereas at the anode, oxygen is provided for oxidative biodegradation. In this paper, it is suggested that the oxygen demand for aerobic *in-situ* soil bioremediation could also be supplied by transport of the oxygen generated by the water oxidation reaction at the surface of the anode in an electrobioremediation process. Several authors have reported different studies using water electrolysis products ( $\text{H}_2$  and  $\text{O}_2$ ) to increase the biodegradation of pollutants in liquid media (Goel and Flora, 2005; Lohner and Tiehm, 2009). However, we did not find studies about the electrokinetic transport of oxygen throughout soil groundwater to supply oxygen for *in-situ* aerobic bioremediation processes. For this purpose, the present work studies the influences of two variables, the soil texture and different voltage gradients, on electrokinetic oxygen transport throughout the water pores of soils. The oxygen concentration and the supply rates are measured, and the feasibility of this technique for possible future bioremediation purposes is discussed.

## 2. Experimental

Bench-scale experiments were performed using the setup shown in Fig. 1. The setup consisted of a horizontal methacrylate column with five compartments: the soil was loaded in the central compartment (20 cm length); two other compartments were located on either side, separated from the central compartment by nylon mesh with a 0.5-mm mesh size, and they served as anode and cathode wells (6 cm length each); finally, additional collector compartments were located at the left and right ends, to collect the liquid overflowing from the respective electrodic wells (6 cm length each).

Graphite electrodes were introduced in the electrodic compartments, connected to the power supply (HQ Power, Gavere, Belgium) so that one of them acted as the anode compartment, and the other one as the cathode compartment. The dimensions of these electrodes, provided by Carbosystem, were 10 cm × 10 cm × 1 cm, so that they had the same cross-section as the fraction of soil to be studied, to have a homogeneous distribution of current lines throughout the soil.

Two different synthetic soils were used to study the influence of the soil texture. The soils used were kaolinite (provided by M. Riesgo Chemical Products, Madrid, Spain) as a model of clay soil, and washed sea sand (provided by Panreac Chemical Products, Madrid, Spain), with a diameter particle between 1 and 2 mm, as a model of sandy soil.

A natural silty soil, collected from the shore of a river, was later used as an example of application in a real soil. This natural soil was subjected to initial thermal treatment (24 h at 105 °C) to remove its natural humidity, after which it was sieved to use only the fraction of soil with a particle size between 20 and 50 μm. The natural silty soil contained 5.5% total organic carbon and 44.9 g kg<sup>-1</sup> total volatile solids (dry basis). It also contained

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