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Removal of oxyfluorfen from spiked soils using electrokinetic fences

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ABSTRACT

In this work, an accidental spill of Fluoxyl (commercial herbicide containing oxyfluorfen) is simulated in a pilot plant with a soil volume of 70 × 50 × 50 cm^3. The transport of Fluoxyl obtained by the free diffusion of pollution and under the application of the electrokinetic fences (EKF) technology are compared in a 34-day treatment. In addition, the temperature, conductivity, and pH are monitored daily. At the end of the experiment, a post-mortem analysis is carried out in order to obtain a 3-D distribution map of the pollutant. The results show that EKF is a good technology to remove oxyfluorfen from the soil without excavation because it is able to attain a fast transfer of oxyfluorfen to the flushing fluid used. After 34 days, the decrease in the concentration of oxyfluofen in the simulated case without any treatment is only 5.5%, whereas when EKF is applied, the removal yield is approximately 63% (60.7% of improvement vs. natural volatilization). Detailed analyses of the experimental data, the 3-D map, and recent literature suggest that the main mechanism involved for the removal is the rapid transfer of oxyfluorfen to the flushing fluid used. The results are also discussed in the context of a previous work in which the same technology has been applied for the removal of the ionic herbicide 2,4-D. A comparison allows sound conclusions to be made for future scale-up studies.

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1. Introduction

Over the last decades, there has been increasing concern about the pollution of soils and that the uncontrolled spreading of pollution may affect water supplies. In particular, the pollution of hazardous wastes is interesting and has been the focus of recent relevant research, both in the characterization of the environmental problem and in the development of promising technologies to avoid the negative consequences of these serious environmental problems.

Among the pollutants that may cause soil pollution, organic pesticides and particularly herbicides are a group of great interest [1]. These products are quite relevant for the economic feasibility of crops, but their influence on the environment is very negative, despite the many efforts made in recent years to produce biodegradable pesticides, whose negative effects disappear within a reasonable period of time.

The development of technology capable of confining the pollution to a constrained zone and avoiding uncontrolled dispersion is an interesting target for the present research on soil remediation [2–10]. However, the main aim is to develop a technology capable of removing the pollutant and allowing the recovery of the soil [2,11–13]. Within the technologies capable of confining pollution, electrokinetic fences (EKF) is perhaps one of the most interesting, at least from a theoretical point of view [11,14].

The EKF technology has been studied for the retention and removal of ionic species [15–19], including pesticides [20–25]. The results published have been promising but also indicate that there is still much work to do in order to completely understand the technology and to be able to use it in an efficient way. As with many other electrokinetic technologies, understanding electrokinetic fences does not only consist of knowing the mechanisms involved (which are already well-known and studied at the lab-scale), but the combination of mechanisms in this type of process needs to be understood [17]. This combination can be complex, especially when studied in systems of large scales (larger than the small soil columns typically shown in most of the papers published in the recent literature) because of the very different time-constants of these processes [26].

One additional disadvantage of this technology is its partial effectiveness for pollutants with a low water solubility [27–29].

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These pollutants need to be transformed into emulsions in order to eliminate them [26,30,31]. This has been shown for the case of oxyfluorfen, i.e., [2-chloro-1-(3-ethoxy-4-nitrophenoxy)-4-(tri fluoromethyl) benzene], which is a diphenyl ether herbicide commonly used in agriculture to control broadleaf and grassy weeds. This herbicide is known to have low water solubility (0.116 mg/L at 20 °C), low vapour pressure (0.026 mPa at 25 °C), high Koc (log-Koc = 3.46–4.13), and high Kow (log Kow = 4.86). This substance is not metabolized in plants and can be slowly digested by microorganisms [32–34]. Hence, oxyfluorfen is an interesting model pollutant, which can give relevant data on soil transport under electrokinetic treatment.

Thus, the goal of this work is to study the use of electrokinetic fences in a pilot plant for soil treatment containing an oxyfluorfen dispersion (Fluoxil 24 EC). The results obtained are compared with those from a blank experiment. Detailed analyses of the experimental data, the 3-D map, and the recent literature suggest that the main mechanism involved for the removal is the rapid transfer of oxyfluorfen to the flushing fluid used.

2. Materials and methods

2.1. Preparation of the polluted soil

Field soil from Toledo (Spain) was used in this study. This soil was characterized by its inertness, low hydraulic conductivity and lack of organic content. The mineralogical composition and the parameters used to classify this soil by the Unified Soil Classification System (USCS) are listed elsewhere [30]. As a model nonpolar and hydrophobic herbicide, 24% Oxyfluorfen (with calcium dodecylbenzenesulfonate as the surfactant, i.e., cleansing agent) was used. The commercial herbicide used was Fluoxil 24 EC (CHEMINOVA AGRO, S.A., Madrid, Spain). The process of soil preparation is important because of the complexity of natural soil. The process was divided into four different stages: (1) Installation of three layers of gravel with different particle sizes for mechanical and drain support; (2) moistening of the soil to 11% (natural water content condition); (3) compaction of the soil in the electrokinetic reactor by compacting layers of a fixed thickness (5 cm) until achieving the natural density of the soil (approximately 1.4 g cm^{-3}); (4) drilling of the electrolyte wells and instrumentation of the plant.

2.2. Experimental setup

The electrokinetic experiments were conducted in an electrokinetic remediation plant consisting of an electrokinetic reactor, a power source, and tanks of electrolyte. The reactor was a methacrylate prism with a soil capacity of 175×10^3 cm^3 (LWH: $70 \times 50 \times 50$ cm^3). The electrodes used for both the anodes and the cathodes were graphite rods with dimensions of $1 \times 1 \times 10$ cm^3, positioned in semipermeable electrolyte wells, using a sequence of alternating electrodes. The cathodic wells were connected to 100 cm^3 sewers to accumulate the fluid transported through the soil and to facilitate sampling. The reactor was designed to separate and collect the fluids through an outlet situated on the side wall of the reactor. To monitor the flux of water and the temperature evolution during the experiment, tensiometers, thermocouples, and rhizon samplers (hereafter, simply "rhizons") were inserted into the soil. Fig. 1 shows a scheme of the well configuration and the instrumentation of the plant. At the top of the soil, a capillary barrier consisting of an approximately 3-cm-wide layer of sand was placed in order to reduce the evaporation of water and the volatilization of herbicide.

2.3. Experimental procedure

Once the plant was completely instrumented, the experimental procedure began with the pollution of the soil (simulating an accidental spill). To do this, an accidental leak of 6.0 dm^{-3} of an

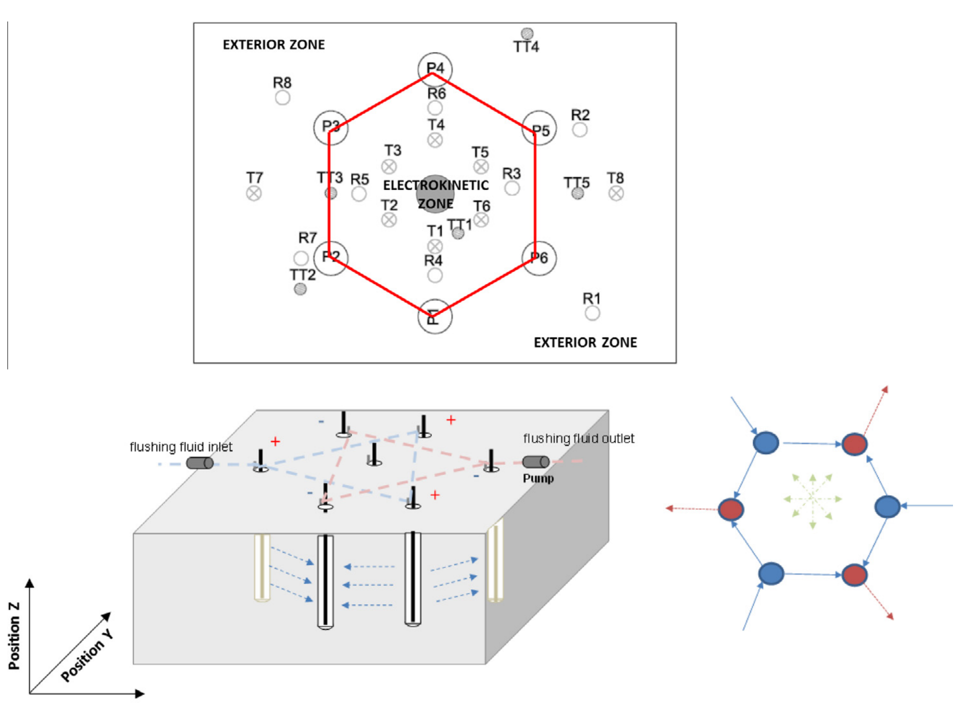

Fig. 1. Scheme of the well configuration and diagram of the instrumentation used in the electrokinetic remediation plant. R_i: Rhizon no. i; TT_i: Thermocouples no. i; T_i: Tensiometers no. i; P_i: Well no. i.

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