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Application of a new electrolyte circulation method for the ex situ electrokinetic bioremediation of a laboratory-prepared pentadecane contaminated kaolinite

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Abstract

Ex situ electrokinetic (EK) bioremediation of a laboratory-prepared pentadecane-contaminated kaolinite was carried out. Extraneous bacteria and ionic nutrients were continuously supplied to the soil specimen by a new electrolyte circulation method, which controlled electrical pH change of electrolyte solution to keep bacterial activity. During the EK bioremediation the anode region showed the highest colony forming unit (CFU) due to electrical attraction between anode and bacteria. Simultaneous increases of CFU and uniform pentadecane removal in most soil regions demonstrated that electro-osmosis as well as electrophoresis affected the bacterial transport in soil. At 3.13 mA/cm^2, increase in soil temperature to above 45 °C inhibited bacterial activity, which caused the decrease of removal efficiency. The removal amount of pentadecane increased with initial pentadecane concentration at the same current densities (0.63 and 1.88 mA/cm^2) because of the increased amount of weakly bound pentadecane onto the soil surface. The highest removal efficiency (77.6%) was obtained at 0.63 mA/cm^2 for 1000 mg/kg pentadecane after 14 days. Consequently, the present methods of EK bioremediation demonstrated superiority over the conventional bioremediation, which had inherent demerits of slow degradation and low removal efficiency.

Keywords: Electrokinetic bioremediation; Pentadecane; Kaolinite; Extraneous bacteria; Electrolyte circulation method

1. Introduction

The widespread usage and storage of petroleum fuels have made petroleum hydrocarbons the most prevalent soil and groundwater contaminants. The treatment of sites contaminated with long chain alkanes and polycyclic aromatic hydrocarbons involved in petroleum fuels has limitations because of their properties such as low volatility, low mobility, low solubility and low degradability [1,2].

Bioremediation has been applied to various contaminated sites for several decades, because it has many advantages such as permanent elimination of waste, cheaper biological system, positive public acceptance, minimum site disruption, risk elimination with long-term liability and combination with other treatment techniques [3]. In a heterogeneous and/or low permeability soil, however, bacteria cannot sufficiently metabolize contaminants due to the transport limitation of bacteria or nutrients, and so an additional management is required [4,5].

When a direct current (dc) field is introduced across soil deposits through inert electrodes, the EK phenomena such as electro-osmosis, electrophoresis, and electromigration cause the transport of various compounds even in a low permeability soil [6–10]. Therefore, application of EK phenomena to bioremediation, namely EK bioremediation, can uniformly and rapidly supply nutrients, electron donors/acceptors, and bacteria to soil. It is generally known that electrophoresis is an important mechanism for bacterial transport [4,5], which depends upon the surface charge density of individual or

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aggregative bacteria. However, few researches on EK bioremediation have been reported.

In this study, ex situ EK bioremediation of a laboratory-prepared pentadecane-contaminated kaolinite was carried out. A new electrolyte circulation method was employed to improve the process efficiency by supplying bacteria and ionic nutrients continuously and preventing electrical pH change. The effects of electric current density and initial pentadecane concentration were investigated on bacterial behavior and removal efficiency. The process feasibility was evaluated for petroleum hydrocarbon-contaminated sites compared to other conventional bioremediation.

2. Materials and methods

2.1. Microorganism and electrolyte composition

The microorganism used in this study was a bacterial consortium consisting of several *Pseudomonas* sp. (L Company, Korea). Electrolyte was prepared to supply the bacterial consortium with ionic nutrients and to increase the electrical conductivity of soil [11]. The composition of electrolyte is as follows (g/l): $Na_2HPO_4{\cdot}12H_2O$ 18.1; KH_2PO_4 4.4; $MgSO_4{\cdot}7H_2O$ 0.2; NH_4Cl 1.0; glucose 10.

2.2. Shaker-flask culture for bacterial growth and pentadecane degradation

Bacterial growth and pentadecane degradation were observed in a 250 ml flask containing 100 ml of culture solution, of which composition was same as the electrolyte including 5% (v/v) pentadecane (3845 mg/l). Three growth media were prepared to avoid the loss of culture solution and pentadecane content in measuring bacterial growth and pentadecane removal each week. The flask cultivation was carried out in a shaking incubator at 30 °C and 150 rpm. The bacterial growth was determined by measuring the optical density at 600 nm using an UV spectrophotometer (Hewlett Packard 8562A, USA). Prior to pentadecane analysis, the bacterial activity was eliminated by adding NaOH into the culture solution to increase pH to above 12. After extracting pentadecane with *n*-hexane at 25 °C and 150 rpm in a shaking incubator, its content was measured by HPLC (details in Section 2.5).

2.3. Specimen preparation

Pentadecane (Sigma, USA), one of the long-chain alkanes contained in diesel fuel, was selected as a model pollutant, because EK bioremediation is appropriate for low volatile pollutants. The model soil used in this EK experiment was kaolinite (Sanchung, Korea) screened with the No. 50 sieve (US standard screen). Kaolinite has a relatively low cation-exchange capacity so that several parameters such as electrical potential gradient, bacterial transport, and soil pH under dc field can be evaluated precisely. After autoclaving of 450 g kaolinite at 121 °C for 15 min the soil water content was controlled to be 25% with deionized water of 150 ml. Initial concentrations of pentadecane artificially contaminated were 1000, 5000, 10,000 and 20,000 mg/kg dry soil. The specimen was loaded into the EK test cell, which was constructed with a horizontal rectangular polyacrylate (20 cm × 4 cm × 4 cm) with stainless steel electrodes coated by platinum immersed in the electrode reservoirs (2.5 cm × 4 cm × 4 cm) as shown in Fig. 1. A peristaltic pump was used to circulate the electrolyte solution at a rate of 4.2 ml min^{-1} to control ionic concentrations and sudden pH change in soil and bioreactor [12,13]. Bioreactor contained 1 l of the electrolyte solution.

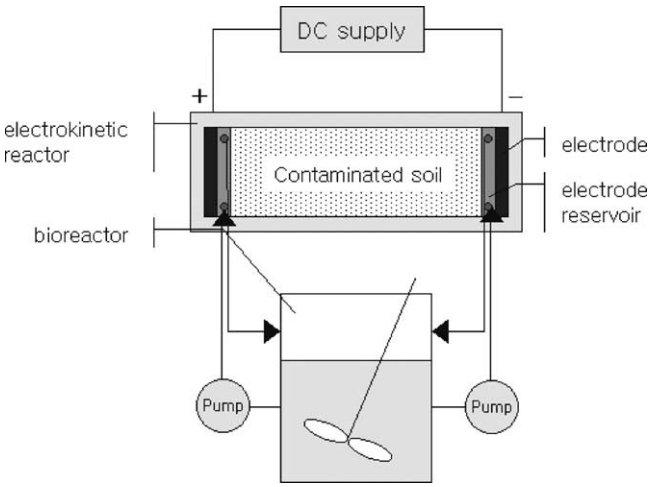

Fig. 1. Schematic diagram of the experimental EK bioremediation.

2.4. EK bioremediation

Before the start-up of EK bioremediation, the bacterial consortium was cultivated up to 0.5 of optical density (600 nm) in a bioreactor. After filling electrode reservoirs with the harvested bacterial solution, the EK bioremediation was operated with a constant current for 2 weeks. The bioreactor was operated at 250–300 rpm and 30 °C.

In a preliminary EK experiment, current densities to increase soil temperature to between 25 and 35 °C at which microorganisms grew rapidly were found to be about 0.31–3.13 mA/cm^2. Therefore, the ranges of current densities applied in the present experiment of ex situ EK bioremediation were between 0.31 and 3.13 mA/cm^2 using a dc power supply with maximum output of 300 V.

The experimental condition is summarized in Table 1: the effect of high current density on the removal efficiency was investigated by experiments 1 and 2. The relationship between contamination level and removal amount was examined at 20,000, 10,000 and 5000 mg/kg pentadecane (experiments 2, 3 and 4). Three different current densities were tested to find an optimum value for 5000 mg/kg pentadecane (experiments 4, 5 and 6). The process feasibility was evaluated