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Electro-bioremediation of contaminated sediment by electrode enhanced capping

Fei Yan [a, *], Danny Reible [b]

[a] Department of Civil and Environmental Engineering, Rice University, Houston, TX 77005-1892, USA
[b] Department of Civil and Environmental Engineering, Texas Tech University, Lubbock, TX 79409-1023, USA

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ABSTRACT

In-situ capping often eliminates or slows natural degradation of hydrocarbon due to the reducing conditions in the sediments. The purpose of this research was to demonstrate a reactive capping technique, an electrode enhanced cap, to produce favorable conditions for hydrocarbon degradation and evaluate this reactive capping technique for contaminated sediment remediation. Two graphite electrodes were placed horizontally at different layers in a cap and connected to external power of 2 V. Redox potentials increased and pH decreased around the anode. Phenanthrene concentration decreased and PAH degradation genes increased in the vicinity of the anode. Phenanthrene concentrations at 0–1 cm sediment beneath the anode decreased to ~50% of initial concentration over ~70 days, while phenanthrene levels in control reactor kept unchanged. A degradation model of electrode enhanced capping was developed to simulate reaction-diffusion processes, and model results show that a reaction-dominated region was created in the vicinity of the anode. Although the degradation dominated region was thin, transport processes in a sediment cap environment are typically sufficiently slow to allow this layer to serve as a permeable reactive barrier for hydrocarbon decontamination.

1. Introduction

Contaminated sediments have become significant problems at many sediment sites both in the USA and other parts of the world. Contaminants detected in sediments include hydrophobic organics as well as metals. In situ capping is a remediation technique to contain contaminated sediments by placing a covering cap made of clean isolating materials at the water–sediment interface. Natural degradation processes for hydrophobic organics may be hindered beneath a cap due to the development of anaerobic conditions and transport processes may ultimately lead to recontamination of near surface sediments.

To encourage degradation of hydrophobic organics after conventional sand capping, it is necessary to maintain conducive conditions to contaminant degradation, e.g., nutrient-sufficient conditions, redox potentials, electron donor or acceptor levels. In organic rich sediments, often the key limitation is providing redox conditions conducive to contaminant transformation.

During recent years, electrochemical technologies have been proposed for sediment, groundwater and soil remediation. Numerous studies have demonstrated the potential effectiveness of electrokinetic remediation in the removal of soil contaminants, primarily due to direct oxidation-reduction at electrodes or induced mobility (Acar and Alshawabkeh, 1993; Franz et al., 2002; Gent et al., 2009; Kim et al., 2011; Lukman et al., 2013; Ng et al., 2014). However, for hydrophobic organic compounds (HOCs) such as polycyclic aromatic hydrocarbons (PAHs) whose mobility is not redox sensitive and with a high tendency to be adsorbed onto soil and sediment, electrokinetic remediation has limited effectiveness without the use of surfactants (Pazos et al., 2010; Hahladakis et al., 2014). Bioremediation is a reasonable alternative to treat soil and sediment polluted by PAHs but the terminal electron acceptor in sediments even a few centimeters below the sediment–water interface is typically iron(III) or sulfate, and this highly reducing conditions are not conducive to PAH degradation. Successful implementation of in-situ bioremediation sometimes requires the presence or injection of electron acceptors into the porous medium. A technology for a continuous introduction of electron acceptors and providing conditions conducive to hydrocarbon oxidation has limited bioremediation of PAH contaminated sediments. Using an electric current may be an alternative option for supplying more

* Corresponding author.
E-mail address: feiyan@utexas.edu (F. Yan).

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favorable electron acceptors and conditions, providing a potentially viable remedial approach (Strycharz et al., 2008; Aulenta et al., 2009; Sun et al., 2010; Zhang et al., 2010; Chun et al., 2013).

In this research, electrode enhanced capping is proposed and evaluated for bioremediation of sediments contaminated with PAHs (Fig. 1). Two areal graphite electrodes are placed horizontally and perpendicular to contaminant transport in the cap and connected to an external power, with low current and power requirements. By water electrolysis, micromolar quantities of oxygen are produced at the anode, making the local sediment environment more oxidizing. Consequently, these redox conditions changes and produced oxygen are expected to stimulate the activity of PAH degrading microbes, and accelerate contaminant biodegradation. The objective of this study is to demonstrate the feasibility of electrode enhanced caps for remediation of PAH contaminated sediment.

2. Materials and methods

2.1. Microcosm setup and operation

Four microcosm reactors were used to investigate redox control and PAH degradation in electrode enhanced caps. The control reactor had no power applied, and the sand caps of the control was 1 mm-sieved concrete sand without any amendment (Riccelli Enterprises, Rush, NY). The external power was connected to the other three reactors through graphite electrodes in the amended sand capping layer. The composition of the amended sand caps was 80 g of siderite, 80 g of calcite, 3.6 g of $NaHCO_3$ and 1 L of 1 mm-sieved concrete sand. It was expected that siderite and calcite would provide long term pH buffering capacity while bicarbonate would provide short term pH buffering capacity. Studies had previously shown that an unbuffered system would exhibit pH excursions which would hinder microbial community development (Sun et al., 2010). The pH in the control cap system was stable without buffering.

The microcosms were filled with 2 mm-sieved sediment (mixture of PAH contaminated Anacostia River sediment and clean sediment, ratio 1:6). The main characteristics of the sediment including pH, redox potential, conductivity, and water content are in supporting materials. The sediment was spiked with a model PAH compound, i.e. phenanthrene, at ~55 μg/g solid concentration. The phenanthrene porewater concentration (as measured by passive sampling, see below) was approximately 30 μg/L. A 13 cm × 6 cm graphite cloth (Carbon Cloth CC6 Plain, Fuel Cell Earth LLC, Wakefield, MA) was placed above the sediment layer as the anode. An amended sand layer of 3.5-cm was laid over the anode, and another graphite cloth was placed in the amended sand layer as the cathode. A 0.5 cm amended sand layer was on top of the cathode. The pictures of microcosms of electrode enhanced caps in this study are available in supporting materials. The sand layers of powered and control reactors were initially saturated with freshwater. Freshwater was made by dissolving 35 g of Instant Ocean® sea salts in 1 L of DI water and diluting by a factor of 150. A multichannel peristaltic pump (Watson Marlow 205s, Watson-Marlow, Inc, Wilmington, MA) was used to pass the freshwater over the surface of the sand in the microcosms.

The graphite electrodes were connected to copper wires by EPO-TEK® 377H (Epoxy Technology, Billerica, MA), which is a graphite filled epoxy designed for electronic industries. The connections between the graphite cloth and copper wires were sealed with WEST SYSTEM 105 epoxy resin and 206 slow hardener (WEST SYSTEM, Bay City, MI) to prevent corrosion. Powered reactors were connected to Extech 382202 Model DC power supplies (Extech Instruments, Waltham, MA). Among the three powered reactors, two of them (CtnCap1 and CtnCap2) were operated in a continuous power mode: an external voltage of ~2 V was applied continuously except during in-situ pH and redox potential measurement; the third powered reactor (IntmtCap) was operated in an intermittent power mode: an external voltage of ~2 V was applied 16 h continuously over a 24-h period. The applied voltage and current for all the three reactors are available in supporting materials.

2.2. pH and ORP measurement

pH was monitored by pH microelectrode MI-405 (Microelectrodes Incorporation, Bedford, NH) and oxidation reduction potential (ORP) was quantified using platinum microelectrode. pH and ORP profiles from water–sand interface to 5.5-cm below were acquired at 0.5 cm intervals.

2.3. Phenanthrene porewater concentration measurement by PDMS-coated fiber

Porewater concentrations of phenanthrene in the sediment and sand were measured by polydimethylsiloxane (PDMS)-coated fibers (Fiberguide, Stirling, NJ), which has a 10-μm coating on a 210-μm core. The capability of the PDMS-coated materials to measure PAH concentrations in porewater has been demonstrated in other studies (Lu et al., 2011).The partition coefficient K_f of phenanthrene

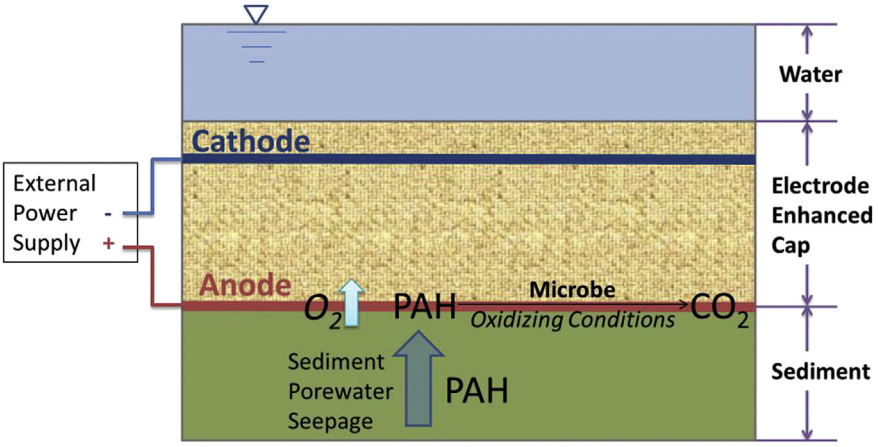

Fig. 1. Conceptual model for an electrode enhanced cap for remediation of PAH contaminated sediment. PAH biodegradation is stimulated by the oxygen produced at the anode.