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University of Vigo. Dept. of Chemical Engineering
Building Isaac Newton. 36310 Vigo. Spain
phone: +34 986 812318 - fax: +34 986 812380
e-mail: erem2007@uvigo.es

ELECTROKINETIC DELIVERY OF NANOSCALE IRON PARTICLES FOR REMEDIATION OF PENTACHLOROPHENOL IN CLAYEY SOIL

Krishna R. Reddy[1], Amid P. Khodadoust[2], and Madhusudhana R. Karri[3]
[1]Professor, [2]Associate Professor and [3]Graduate Research Assistant
University of Illinois at Chicago, 842 West Taylor Street, Chicago, Illinois 60607, USA
Tel: 312-996-4755; Fax: 312-996-2426; E-mail: kreddy@uic.edu

Several researchers have synthesized different types of nanomaterials for a wide range of engineering applications. However, only recently nanoscale iron particles (NIPs) have received the attention of environmental professionals to remediate contaminated sites. This paper investigates: (1) the reactivity of NIPs to promote the reductive degradation of pentachlorophenol (PCP) in clayey soils, and (2) potential to deliver NIPs into PCP-contaminated clayey soils using electrokinetics and resulting PCP reduction efficiency.

Kaolin was used as a model low permeability clayey soil and was artificially spiked with PCP at initial concentration of 500 mg per Kg of dry soil (mg/Kg) or 1000 mg/Kg. PCP was chosen because it is one of the common contaminants found at former wood preserving contaminated sites. To spike the soil, hexane was used to dissolve solid PCP, and this hexane-PCP mixture was slowly added to dry kaolin soil. The mixtures were stirred with stainless steel spoons within glass beakers and then placed beneath a ventilation hood for nearly a week until the hexane completely evaporated and the contaminated soil was dry. The dry soil was then mixed with 35% deionized water to simulate typical field moisture conditions. The NIPs used for this study were produced using the the patented method by Uegami et al. (US2003/0217974A1) and consisted of an elemental iron core (α-Fe) and a magnetite shell (Fe_3O_3) in approximately same amounts by weight. The aqueous NIP suspension with 25.6 wt.% solid concentration had density of 1.27 g/mL. The average particle size and surface area of NIPs was 70 nm (0.07 μm) and 28.8 m^2/g, respectively.

A series of laboratory batch experiments was conducted on PCP-contaminated kaolin with different contact time (1, 2, 8, 12, 24 and 48h) and different NIPs concentrations (1, 2, 4, 5, 8, 10, 20, 40, 60, 80 and 100 g/L) to investigate the reactivity and optimum concentration of NIPs. These experiments were conducted in 40-mL glass vials containing 25 mL of NIPs suspension of known concentration (5, 10 and 20 g/L) and 5 g of PCP spiked kaolin soil (with PCP concentration of 1000 mg/Kg). The soil-NIPs suspensions were shaken for 48 hours. The aqueous suspension was centrifuged at 4000 rpm for 30 min to separate the solids from the liquid. The supernatant and the residual soil were analyzed for PCP concentration using GC-MS. These results showed that 80 to 98% PCP was removed from the soil within an hour, but PCP reduction was increased from 50 to 78% at 1h to 40 to 90% at 24 h reaction time for different NIP concentrations. There was no significant effect of NIPs concentration on the PCP removal, but the amount of PCP reduction increased with increase concentration of NIPs with 30% at 1 g/L to 98% at 100 g/L. There appears to be an optimal NIPs concentration beyond which benefits are diminished.

A series of electrokinetic experiments was conducted to investigate electrokinetic delivery of NIPs and resulting PCP reduction efficiency. The testing equipment and the procedure were the same as that used in previous studies at University of Illinois at Chicago (UIC). Kaolin spiked with PCP with an initial concentration of 500 mg/Kg was tested under different NIP concentrations, voltage gradients, operating duration, and enhancement conditions as summarized in Table 1. Except for the control test, the anode reservoir was filled with NIPs suspension and recirculated with a pump, while the cathode reservoir was filled with deionized water. During the application of electric potential, current and the electroosmotic flow were recorded. At the end of each test, aqueous samples from the electrodes and dissected soil sections were analyzed for pH, iron and PCP.

Table 1. Testing Program to Investigate Electrokinetic Delivery of NIPs

Test Date	Test Designation	Voltage Gradient (VDC/cm)	Anode Flushing Solution	Test Duration (Hours)	Pore Volumes
March 31, 2003-April 18, 2003	NIP0	1.0	Distilled Water	427	1.2
	NIP1	1.0	5 g/L NIP	427	2.1
	NIP2	1.0	10 g/L NIP	427	2.2
August 28, 2003- October 7, 2003	NIP3	2.0	5 g/L NIP	937	0.9
	NIP4	2.0	10 g/L NIP	936	1.5
	NIP5	1.0	0.5% Tween 80 + 5 g/L NIP	936	2.4
	NIP6	1.0	5% Ethyl Alcohol + 5 g/L NIP	936	2.5

The test results showed that the current increased rapidly during the first few hours, then decreased over a period of time. The measured electroosmotic flow varied depending on the test conditions as shown in Table 1. Very low amounts of iron and PCP were detected in the effluent. The dissected soil sections revealed that soil pH decreased in the first four sections from the anode but was the highest in the section closer to the cathode. The iron concentrations in soil increased from the anode to the cathode, indicating the delivery of NIPs into the soil using electrokinetics. Visual observations also revealed the presence of iron particles within the first two sections from the anode. A low concentration of PCP was observed in the first section away from the anode that increased up to the third section and then further decreased towards the cathode end, indicating the migration of PCP occurring from anode to cathode. The amount of PCP reduction by NIPs was calculated based on the initial mass in the soil, the mass removed in the effluent, and the mass remaining in the soil and it ranged from 100% near the anode to 5% near the cathode. The enhanced electrokinetic experiments results showed that high electroosmotic flow was obtained in the cosolvent enhanced system (2.5 pore volume) as compared to surfactant enhanced system (2.4 pore volume) resulting in the increased injection of nanoiron into the soil. Further it was also evaluated that the transport and dispersion of nanoiron particle was more uniform in surfactant enhanced system as compared to cosolvent enhanced system.

Overall, the results from this study revealed that the extent of delivery and reactivity of NIPs were limited by passivation and aggregation of NIPs under the oxygenated and low pH conditions that exist at the anode as well as complex geochemical reactions occurring simultaneously at different rates. The NIPs transport may be affected by probable dissolution of substrate minerals and possible precipitation of secondary solids as well as changes in the surface charge of all solids as the solution composition changes. Parameters controlling these reactions include pH, Eh, solution composition, and solid (both NIPs and substrate) composition and structure. Besides these considerations, the system parameters such as voltage gradient, mode of voltage gradient application (pulsed versus continuous), and pH control at the anode and cathode should be optimized.