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A NEW APPROACH TO SOIL REMEDIATION: COUPLING NANOTECHNOLOGY WITH ELECTRICALLY INDUCED PARTICLE TRANSPORT (ELECTROKINETICS)

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ABSTRACT

Traditional remediation technologies for Persistent Organic Pollutants (POP) such as Polychlorinated Biphenyls (PCBs) have serious limitations and high costs. Zero valent iron nanoparticles (nZVI) represent a new generation of environmental remediation technologies that could provide cost-effective solutions. However, the limited transport of nZVI through soils has been identified to be a major drawback for *in situ* applications. The main research objective of this study is to find out if coupling electrokinetics and reactive iron nanoparticles can be an effective method for treating PCBs contaminated soils, through electrically induced nanoparticle transport. To understand the fundamental aspects of the coupled technology the authors suggest dividing the overall problem into three submodels (transport of nanoparticles by electroosmosis and by electrophoresis and degradation of PCB) which are then integrated into a macroscale transport/degradation numerical model. This paper presents the experimental approach suggested for each submodel under study. Testing this innovative solution using the suggested approach will allow for a deeper understanding of both the electrokinetically enhanced transport of nZVI and PCBs dechlorination.

Keywords: Electrokinetic remediation; Zero Valent Iron nanoparticles (nZVI); coupled technologies; Persistent Organic Pollutants

INTRODUCTION

Soil contamination is a major environmental issue worldwide, as a result of mining, industrial and urban activities over the past two centuries. Of special concern are Persistent Organic Pollutants (POPs), namely Polychlorinated Biphenyls (PCBs). This family of synthetic organic compounds comprises 209 congeners that differ in chlorine number and position and with unique chemical properties. PCBs are very persistent to degradation and hazardous to the biosphere [1]. Estimates of total PCBs production vary between 1.2 and 2 million t, with some of the most detailed data indicating a total global production of approximately 1.3 million t over the period from 1929 to 1993 [2]. From this cumulative global production, about 440 to 92,000 t is estimated to have been emitted into the environment [2, 3].

In the United States, 350 of the 1,290 Superfund Sites are contaminated with PCBs [4]. In Canada, 148 sites are contaminated with PCBs in the total inventoried 20,762 federal sites [5]. In European countries, an estimate points to 242,000 contaminated sites and about 2.4% are contaminated with chlorinated hydrocarbons [6]. An inventory on atmospheric deposition in background surface soil, estimates a global total PCBs soil burden of 21,000 t [7].

The most frequent soil remediation technologies used for PCBs are “dig and dump” and also “dig and incinerate”. It is of utmost importance to find cost-effective technologies to remove PCBs from soils and sediments. This has been a challenging task for decades, since traditional methods like excavation/landfilling and incineration are expensive and imply pollution transfer.

Zero valent iron nanoparticles (nZVI) are very effective in the degradation of a wide variety of organic soil contaminants, such as chlorinated organic solvents, organochlorine pesticides and PCBs [8, 9]. The principle behind nZVI is reductive dehalogenation, in which iron promotes the removal of chlorine from the pollutant molecules, degrading them into harmless by-products. Zero valent iron nanoparticles are traditionally injected under pressure and/or by gravity to the subsurface where treatment is needed. However, transport of nZVI is normally limited by their aggregation and settling [10] with mobility in the subsurface being normally less than a few meters.

Since the late 1980s electrokinetic (EK) remediation has been successfully used to treat different types of soils [11-16] and waste materials [17-22]. Although electrokinetic removal of heavy metals from soils is one of the most studied processes [23], electroremediation has also been applied to organic contaminants, such as herbicides [24], creosote [25], chlorinated solvents [26-27], petroleum hydrocarbons [28-29], phenol [30] and PAHs [31-33].

The method uses a low-level direct current as the ‘‘cleaning agent’’, to remove matrix contaminants. Several transport mechanisms (electroosmosis, electromigration and electrophoresis) and electrochemical reactions (electrolysis and electrodeposition) are induced by this method [11]. The contaminants are then moved towards one of the electrode compartments where they become concentrated. The general principle of the electrokinetic process is presented elsewhere and several authors have critically reviewed its state of knowledge [23, 34].

This work proposes the use of electrokinetics to enhance nZVI mobility and improve standard soil nano remediation. The concept behind this research is to create electric fields to transport nanoparticles through the soil, so that they come into contact with the pollutants.

ZERO VALENT IRON NANOPARTICLES

A nanoparticle – a collection of tens to thousands of atoms measuring about 1-100 nm in aggregate diameter and characterized by crystalline shapes and lattice structures – is the most basic structure in nanotechnology [35].

Nanoscale iron particles have large surface areas, high surface area-to-volume ratios and a large fraction of stepped surface [36]. Such properties combine with a unique structure and zero valence to make nano-sized metals extremely reactive [35, 36].

In general, reactions between chlorinated organic compounds ($C_xH_yCl_z$) and iron in aqueous solutions can be expressed by the following reaction [37]:

$$C_xH_yCl_z + zH^+ + zFe^0 \rightarrow C_xH_{y+z} + zFe^{2+} + zCl^- \quad (1)$$

in which iron acts as a reductant (electron donor) for the removal of chlorine. This reaction is similar to the process occurring during iron corrosion, with the beneficial effects of transforming chlorinated pollutants. Reductive dechlorination of PCBs by nanoscale zero-valent iron was first reported by Zhang and Wang [38].The iron nanoparticles exhibit characteristics of both iron oxides (e.g. as a sorbent) and metallic iron (e.g. as a reductant) [39]. Table 1 presents a summary on the review of some of the most relevant studies for the remediation of contaminated matrices with nZVI.

Nanoparticles provide more flexibility for *in situ* applications than granular iron and can remain reactive towards contaminants in soil and water for extended periods of time (> 4-8 weeks) [36]. The nanoparticle–water slurry can be injected under pressure and/or by gravity to the contaminated plume where treatment is needed and nanoparticles can be transported effectively by the flow of groundwater.

COUPLING NANOTECHNOLOGY WITH ELECTROKINETICS

The natural tendency of nZVI to aggregate and/or bind with soil grains is currently a limitation to the successful delivery of nZVI into porous media formations and into low permeability soils. This work proposes the use of electrokinetics (EK) as a way to overcome this limitation. In this coupled technique the role of EK would be quite the opposite of the traditional one: instead of aiming at getting the contaminants out, the electric driving force is used to get nanoparticles into the soil.

Table 1. Summary of studies on zero valent iron nanoparticles.

Matrix	Contaminant	Nanoparticle	% Removal	Reference
Aqueous solutions	Cr(VI) and Pb(II)	Zero valent iron (ZVI)	-	[40]
Groundwater (field case)	Trichloroethene and other chlorinated hydrocarbons	Bimetallic Fe/Pd nanoparticles	96%	[41]
Water/Methanol Solution	PCBs	ZVI nanoparticles and palladized microscale ZVI	-	[42]
Water	TCE and PCBs	Starch-stabilized bimetallic (Fe-Pd) nanoparticles	80%	[43]
Spiked water	As (III)	Zero valent iron (ZVI)	100%	[44]
Groundwater (field case)	VOC	Zero valent iron (ZVI)	65-99%	[45]
Soil	PCBs	Zero valent iron (ZVI)	95%	[46]
Liquid waste effluent	Uranium	Zero valent iron (ZVI)	98,5%	[47]
Spiked water	Hexachlorobenzene	Zero valent iron (ZVI)	-	[48]

Advantages of this approach are that PCBs are degraded into simpler, non-hazardous molecules and remaining nZVI nanoparticles will eventually aggregate and settle, being incorporated in the soil. Contaminants are therefore destroyed *in situ*, not extracted, avoiding the need for further treatment. Transfer of pollution between environmental compartments is avoided, resulting in a more sustainable approach.

Theoretically it is possible to use electric fields to move nanoparticles through the soil to where they are wanted. Transport mechanisms expected to be relevant in this transport are electrophoresis and electroosmosis. Some work has already been done to test this possibility such as the one conducted by Pamukcu *et al.* [49], in which polymer coated nanoparticle are transported in kaolin by electrophoresis. Jones *et al.* [50] also found that nZVI could be transported through fine grained sand with rates comparable to those predicted by electrokinetic theory. More recently, Reddy *et al.* [51] studied the transport and reactivity of lactate modified nanoscale iron particles in kaolin under applied electric potential and found that electrokinetics can enhance delivery of nanoscale iron particles in low permeability soils. This has been confirmed by yet another study [10] where the transport of nZVI under different electric potentials in low permeable kaolin soil was found to increase with higher voltage gradients.

These preliminary studies use model soils spiked with contaminants, and are far from the real conditions found in the field. Additionally, although electrokinetic remediation has been used quite extensively for different pollutants, it has never been used to extract PCBs from soils nor coupling electrokinetics and nanoparticles to remediate PCBs contaminated soils and sediments has ever been reported. Coupling the two technologies, EK and nZVI, is thus a promising technology but still at a very early stage of development. In fact, basic aspects of this complex problem still remain unknown (such as *how are PCBs attached to the soil* and *how this influences degradation/dechlorination rate; what are the best operational conditions; how long will the nanoparticles be reactive; what is the influence of different soil conditions).* Perhaps one of the most relevant question is how accurately can nZVI be moved *to a specific location* under real soil conditions using electric fields and how long will this take.

EXPERIMENTAL APPROACH

To investigate the fundamental aspects of nZVI coupled with EK it is necessary to describe the behavior of the PCB-nZVI-soil system, both quantitative and qualitative. Due to its complexity the methodological approach proposed here is to divide the problem into smaller, simpler parts, solve each part and integrate them into a global model. It is assumed that each part of the problem is independent from the remaining parts, and that the total equals the sum of its parts. The following 3 sub-models need studying:

- Transport of nZVI by electrophoresis
- Transport of nZVI by electroosmosis
- PCB degradation

The experimental approach suggested for formulating the three sub-models is presented next.

To investigate the electrophoretic transport of nanoparticles a laboratory cell similar to the one described in Pamukcu *et al.* [49] can be used. Different materials should be tested, starting with surrogate porous media, such as glass beads, then sand and pure clay (kaolinite), before using actual soil. This way the transport of nZVI in oxidizing/reducing environments can be understood when no interactions with the substrate occur. After establishing the performance nZVI transport under the electric filed (with and without pH control) and determining a mobility index, the experiments should go one step further to include mixed substrates and real soils.

For the study of the electro-osmosis transport mechanism the laboratory cell developed at Department of Earth Sciences of Utrecht University (DES-UU) is recommended, after adapting to allow for monitoring of input and output quantities of nZVI. This cell allows to measure the magnitude of the electro-osmosis effect in water saturated, clayey soil samples and to study the transport of nanoparticles through the soil by means of electroosmosis.

For the third sub-model, PCB degradation, the strategy consists in conducting batch extraction procedures using real contaminated soils, and determine reaction kinetics under different experimental conditions (e.g. concentration of nZVI, electrolyte, current density, duration) in order to optimize PCB dechlorination. The use of surfactants can enhance PCB desorption from soils, and this should also be considered at this stage.

GLOBAL MODEL

Integration of the individual processes can be accomplished by developing a macro-scale transport/degradation numerical model which calculates the concentration of PCB and nZVI at each location with time.

Suggested methodology is to use experimental data obtained in each sub-model to incorporate into existing models after adaptation. Since mass transport in a porous media (without the application of electric fields) is already extensively addressed, work should focus on transport processes related to the electric field, namely electro-osmosis and electromigration/electrophoresis. For this effect the numerical code developed at DES-UU and TU-Delft to simulate chemical and induced electro-osmotic transport in clayey soils can be adapted. Currently this model includes geochemical processes and contaminant transport during electro-remediation of Polycyclic Aromatic Hydrocarbons (PAH) contaminated clays. It is currently being upgraded to include active electro-osmosis, clay weathering and contaminant-mobilization. A step further is to integrate electro-kinetic transport of nanoparticles and PCB-degradation kinetics.

This macroscale model will provide a useful tool in the prediction of future applications of nZVI+electrokinetics to remediate other chlorinated hydrocarbons contaminated soils.

CONCLUDING REMARKS

Using electrokinetics in conjunction with nZVI is an innovative solution, since the pollutants will not be extracted from the soil (traditional outcome in electrokinetics) but the electric fields will instead be the driving force behind the movement of nanoparticles deep into the subsurface, where they degrade organic contaminants into harmless by-products. This is conceptually a waste-free technique: existing pollutants are not extracted but *in-situ* destroyed, and iron nanoparticles will eventually aggregate and settle, being incorporated in the soil. This approach avoids pollution transfer between environmental compartments and represents an integrated approach to recover PCB contaminated soils. There is an expected higher efficiency of the coupled techniques when compared to the individual ones, as well as a reduction in the remediation costs when incineration and other *ex situ* treatments are considered.

With this new approach, remediation of low permeability soils will become possible. Even when hydraulic transport is reduced, nanoparticles are still transported by electric fields, avoiding common issues of agglomeration and settlement of these particles.

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