



Electromagnetic induction of nanoscale zerovalent iron particles accelerates the degradation of chlorinated dense non-aqueous phase liquid: Proof of concept



Tanapon Phenrat ^{a, b, *}, Itsaraphong Kumloet ^{a, b}

^a Research Unit for Integrated Natural Resources Remediation and Reclamation (IN3R), Department of Civil Engineering, Faculty of Engineering, Naresuan University, Phitsanulok, 65000, Thailand

^b Center of Excellence for Sustainability of Health, Environment, and Industry (SHEI), Faculty of Engineering, Naresuan University, Phitsanulok, 65000, Thailand

ARTICLE INFO

Article history:

Received 5 August 2016

Received in revised form

12 October 2016

Accepted 13 October 2016

Available online 14 October 2016

Keywords:

Polyelectrolyte-modified nanoscale zerovalent iron (PSS-modified NZVI)
Electromagnetic field (EMF)
Dechlorination
Dense non-aqueous phase liquid (DNAPL)

ABSTRACT

In this study, a novel electromagnetically enhanced treatment concept is proposed for in situ remediation of a source zone of chlorinated dense non-aqueous phase liquid (DNAPL) that is slowly dissolved, causing contaminated groundwater for centuries. Here, we used polystyrene sulfonate (PSS)-modified nanoscale zerovalent iron (NZVI) particles (ferromagnetic) in combination with a low frequency (LF) (150 kHz) AC electromagnetic field (EMF) to accelerate the degradation of the DNAPLs via enhanced dissolution and reductive dechlorination. Trichloroethylene (TCE) and tetrachloroethylene (PCE) were used in a bench-scaled evaluation. The PSS-modified NZVI successfully targeted the DNAPL/water interface, as evidenced by the Pickering emulsion formation. Dechlorination of TCE- and PCE-DNAPL was measured by quantifying the by-product formation (acetylene, ethene, and ethane). Without magnetic induction heating (MIH) by LF EMF, PSS-modified NZVI transformed TCE- and PCE-DNAPL to ethene and ethane at the rate constants of 12.19×10^{-3} and $1.00 \times 10^{-3} \mu\text{mol/h/m}^2$, respectively, following pseudo zero-order reactions. However, four MIH cycles of PSS-NZVI increased the temperature up to 87 °C and increased the rate constants of TCE-DNAPL and PCE-DNAPL up to 14.58 and 58.01 times, respectively, in comparison to the dechlorination rate without MIH. Theoretical analysis suggested that the MIH of the PSS-modified NZVI enhanced the dechlorination of TCE- and PCE-DNAPL via the combination of the enhanced thermal dissolution of DNAPL, the effect of increasing the temperature on the rate constant (the Arrhenius equation), and the accelerated NZVI corrosion. Nevertheless, the effect of the Arrhenius equation was dominant. For the first time, this proof-of-concept study reveals the potential for using polyelectrolyte-modified NZVI coupled with LF EMF as a combined remediation technique for increasing the rate and completeness of in situ chlorinated DNAPL source remediation.

© 2016 Elsevier Ltd. All rights reserved.

Abbreviations: AC, Alternate Current; CVOCs, Chlorinated Volatile Organic Compounds; EMF, Electromagnetic Field; EMFG, Electromagnetic Field Generator; ERH, Electrical Resistance Heating; IR, Infrared; LF, Low Frequency; DNAPL, Dense Non-aqueous Phase Liquid; MIH, Magnetic Induction Heating; MWH, Microwave Heating; NZVI, Nanoscale Zerovalent Iron; PCE, Tetrachloroethylene; PSS, Polystyrene Sulfonate; RFH, Radio Frequency Heating; RNIP, Reactive Nano Iron Particle; TCE, Trichloroethylene; TEM, Transmission Electron Microscope; VSM, Vibrational Sample Magnetometer.

* Corresponding author. Research Unit for Integrated Natural Resources Remediation and Reclamation (IN3R), Department of Civil Engineering, Faculty of Engineering, Naresuan University, Phitsanulok, 65000, Thailand.

E-mail address: pomphenrat@gmail.com (T. Phenrat).

1. Introduction

Chlorinated dense non-aqueous phase liquids (DNAPLs) entrapped in the subsurface as a result of either an accidental or intentional spill slowly dissolve, generating a toxic plume of contaminated groundwater. In addition, the DNAPLs persist as a long-term source of groundwater contamination, making the restoration costly and technically challenging and making it difficult to meet the cleanup targets in a reasonable amount of time through conventional remediation techniques (Fagerlund et al., 2007; Illangasekare et al., 1995; ITRC, 2002; Leeson et al., 2003; US.EPA, 2003). Nanoscale zerovalent iron (NZVI) particles are capable of the reductive transformation of chlorinated organics into

Nomenclature

ΔT	the increasing temperature of the suspension
ΔT_{Max}	the maximum induced temperature
ΔU	the area under hysteresis curves
E_a	the activation energy of dechlorination
EF_T	the total enhancement factor
EF_A	the enhancement factor according to the Arrhenius equation
EF_D	the thermally enhanced dissolution factor (EF_D)
EF_C	the accelerated corrosion enhancement factor
f	frequency of EMF
k_H	the heat induction rate constant
k_{MIH}	the dechlorination rate constants with MIH, theoretically governed only by the effect of the Arrhenius equation
$k_{No MIH}$	the dechlorination rate constants without MIH
k_{SA}	the pseudo surface area normalized zero-order by-product formation rate constants
P_{MIH}	the amount of power dissipated by MIH
R	the ideal gas constant (8.314 J/K*mole)
S_{TMIH}	the solubility of TCE DNAPL and PCE DNAPL at T_{MIH}
$S_{T No MIH}$	the solubility of TCE DNAPL and PCE DNAPL at $T_{No MIH}$
T_{MIH}	the average temperatures of the reactors as a result of MIH (K)
$T_{No MIH}$	the room temperature (K)

more environmentally benign by-products, including acetylene, ethane, and ethene (He et al., 2007; Johnson et al., 2013; Kocur et al., 2015; O'Carroll et al., 2013; Phenrat et al., 2009b, 2015, 2010b; Sakulchaicharoen et al., 2010; Tratnyek and Johnson, 2006; Zhang et al., 1998; Zhao et al., 2016). Due to their small size, novel DNAPL source-zone removal using NZVI has been pursued by researchers (Bishop et al., 2010; Fagerlund et al., 2012; He et al., 2010; Henn and Waddill, 2006; Phenrat et al., 2011a, 2011b; Quinn et al., 2005; Saleh et al., 2005, 2007; Su et al., 2012; Taghavy et al., 2010; Zhan et al., 2009) with the aim of speeding up groundwater restoration via source-zone elimination.

One of the conceptual models of active and selective DNAPL source-zone removal involves the delivery of polymer-modified NZVI to target DNAPL sources and subsequently perform reductive dechlorination. This original idea was proposed by Saleh and coworkers (Saleh et al., 2005) who used NZVI modified by an amphiphilic triblock copolymer that contained polyanionic blocks to stabilize NZVI in suspension and a hydrophobic poly (methyl methacrylate) block to drive NZVI adsorption to the NAPL/water interface. They revealed the potential for NAPL targeting by demonstrating that the triblock copolymer-modified NZVI adsorbed strongly on NAPL/water interfaces via forming a Pickering emulsion. Recently, Phenrat et al. (2011a) demonstrated that NZVI modified by olefin-maleic acid copolymer could target entrapped NAPL source zones at various NAPL saturations in two dimensional, flow-through porous media more relevant to the field conditions.

However, delivering NZVI to the chlorinated DNAPL/water interface appears to be insufficient for effective DNAPL source-zone removal. Although at the DNAPL/water interface, NZVI increased the efficient use of Fe^0 for reductive dechlorination (Liu et al., 2007), under groundwater flow conditions, DNAPL dissolution to the aqueous phase is a rate-limited step of dechlorination (Miller et al., 1990; Powers et al., 1992; Saba and Illangasekare, 2000). This is

because the reductive dechlorination reaction is surface mediated; therefore, contaminants must be dissolved from the DNAPL to transport and adsorb onto the NZVI surface for electron transference. For this reason, the rate of DNAPL degradation is limited by the rate of mass transfer to the aqueous phase (Berge and Ramsburg, 2010; Fagerlund et al., 2012; Phenrat et al., 2010b, 2016; Taghavy et al., 2010; Zhang et al., 2011). Rather, the NZVI reacts with water to form H_2 , which increases the amount of NZVI required for remediation (Berge and Ramsburg, 2010; Liu et al., 2007). Therefore, any action to enhance the DNAPL dissolution can speed the reaction rate and improve the electron efficiency of the remediation, demonstrating the beneficial use of thermal enhanced dissolution together with reductive dechlorination.

Combining thermal dissolution with reductive dechlorination can be achieved by electrical heating, such as electrical resistance heating (ERH) or electromagnetic (EM) heating, such as radio frequency heating (RFH) and microwave heating (MWH). The ERH delivers an electric current to the subsurface and converts electrical energy to thermal energy due to the resistance of the subsurface material to the flow of electricity (ohmic heating). Recently, Truex et al. combined moderate-temperature subsurface ERH (up to 50 °C) with in situ zerovalent iron (ZVI) treatment on a field scale using an average power of 450 kWh per day for a 58 m³ treated area. They reported acceleration of TCE degradation at four to eight times, presumably due to increasing both the reaction and TCE dissolution from DNAPL (Truex et al., 2011). In contrast, EM heating, such as RFH and MWH, utilizes the conversion of EM energy into thermal energy through the interaction between the EM field and the atoms or molecules present in the irradiated material. Thus, the interaction is governed by the nature of the irradiated material and radiation frequency. Moreover, RFH and MWH utilize frequencies as high as 500 kHz to 500 MHz (Lowe et al., 1999; Price et al., 1999) and 500 MHz to 500 GHz, respectively (Falciglia et al., 2013; Lowe et al., 1999). Heating of nonmagnetic aquifer materials arises from conduction losses and dielectric losses. Both electrical heating and EM heating are used in field-scale applications for oil recovery (Bera and Babadagli, 2015; Vermeulen and McGee, 2000) and soil remediation (Beyke and Fleming, 2005; Gavaskar et al., 2007; Price et al., 1999; Vermeulen and McGee, 2000). Further, ERH is reported to remove chlorinated volatile organic compounds (CVOCs) by as much as 98% in water-saturated soil after 175 days of a field-scale test (Beyke and Fleming, 2005). Similarly, RFH was also reported to achieve a similar removal efficiency in water-saturated soil of a field-scale test (Smith and Hinchey, 1993).

While only conduction loss and dielectric loss are majorly responsible for the RFH of soil, magnetic loss is another heating mechanism capable of operating at a lower frequency and thus is theoretically capable of decreasing the transmission losses and the capital cost of EM field (EMF) generation equipment. The EM induction heating (MIH) is through hysteresis loss of magnetic materials due to the irreversible magnetization in an EMF. At a particular frequency (f) of EMF, the amount of power dissipated by MIH (P_{MIH}) (in Watts) can be expressed as (Li et al., 2010):

$$P_{MIH} = f\Delta U, \quad (1)$$

where ΔU is the area under hysteresis curves, governed by characteristics of magnetic materials. The degree of irreversibility, ΔU , is related to the amount of energy dissipation on the reversal of the field. The MIH at a low frequency (LF) EMF (30–300 kHz) is substantially studied in medicine for thermal treatments, such as magnetic-assisted hyperthermia, where functionalized magnetic nanoparticles, such as magnetite target tumor cells, are then heated by applied LF EMF to kill cancer cells (Bañobre-López et al., 2013; Lim et al., 2007).

Download English Version:

<https://daneshyari.com/en/article/5759198>

Download Persian Version:

<https://daneshyari.com/article/5759198>

[Daneshyari.com](https://daneshyari.com)