



Transport of carbon colloid supported nanoscale zero-valent iron in saturated porous media



Jan Busch ^{a,*}, Tobias Meißner ^{b,1}, Annegret Potthoff ^{b,2}, Sascha E. Oswald ^{a,3}

^a University of Potsdam, Institute of Earth and Environmental Science, Karl-Liebknecht-Str. 24-25, 14476 Potsdam-Golm, Germany

^b Fraunhofer IKTS, Winterbergstraße 28, 01277 Dresden, Germany

ARTICLE INFO

Article history:

Received 20 September 2013

Received in revised form 15 May 2014

Accepted 20 May 2014

Available online 27 May 2014

Keywords:

Nanoscale zero-valent iron (nZVI)

Nanomaterial

Carbo-Iron®

Colloid transport

Mobility

In-situ remediation

ABSTRACT

Injection of nanoscale zero-valent iron (nZVI) has recently gained great interest as emerging technology for in-situ remediation of chlorinated organic compounds from groundwater systems. Zero-valent iron (ZVI) is able to reduce organic compounds and to render it to less harmful substances. The use of nanoscale particles instead of granular or microscale particles can increase dechlorination rates by orders of magnitude due to its high surface area. However, classical nZVI appears to be hampered in its environmental application by its limited mobility. One approach is colloid supported transport of nZVI, where the nZVI gets transported by a mobile colloid. In this study transport properties of activated carbon colloid supported nZVI (c-nZVI; $d_{50} = 2.4 \mu\text{m}$) are investigated in column tests using columns of 40 cm length, which were filled with porous media. A suspension was pumped through the column under different physicochemical conditions (addition of a polyanionic stabilizer and changes in pH and ionic strength). Highest observed breakthrough was 62% of the injected concentration in glass beads with addition of stabilizer. Addition of mono- and bivalent salt, e.g. more than 0.5 mM/L CaCl_2 , can decrease mobility and changes in pH to values below six can inhibit mobility at all. Measurements of colloid sizes and zeta potentials show changes in the mean particle size by a factor of ten and an increase of zeta potential from -62 mV to -80 mV during the transport experiment. However, results suggest potential applicability of c-nZVI under field conditions.

© 2014 Elsevier B.V. All rights reserved.

1. Introduction

Contamination of groundwater with organic and inorganic pollutants, such as chlorinated hydrocarbons, fuels and metallic ions, is a threat for environmental and human health. Remediation of groundwater resources has been a subject to science for several decades, but conventional remediation and treatment technologies, such as pump-and-treat, have shown

limited effectiveness in reducing contamination and are expensive (Mueller and Nowack, 2010; Savage and Diallo, 2005). One new strategy to address this issue is the application of nanotechnology, which can be considered as a beneficial technology for sustainable groundwater management (Grieger et al., 2010). Several classes of functional nanomaterials have been developed within the last years such as (1) metal containing nanoparticles, (2) carbonaceous nanomaterials, (3) zeolites, and (4) dendrimers. Different kinds of particles show a variety of physicochemical properties and are therefore interesting for different applications (Savage and Diallo, 2005). The first class of metal containing nanoparticles shows high potential for groundwater and soil remediation and could probably be applied alongside to classical methods or could be installed as in situ permeable reactive barrier. Especially using nanoscale zero-valent iron (nZVI) as a reactive agent seems to be a

* Corresponding author. Tel.: +49 331 977 2649; fax: +49 331 977 2092.

E-mail addresses: jan.busch@uni-potsdam.de (J. Busch),

tobias.meissner@ikts.fraunhofer.de (T. Meißner),

annegret.potthoff@ikts.fraunhofer.de (A. Potthoff),

sascha.oswald@uni-potsdam.de (S.E. Oswald).

¹ Tel.: +49 351 2553 7872; fax: +49 351 2554 147.

² Tel.: +49 351 2553 7761; fax: +49 351 2554 147.

³ Tel.: +49 331 977 2675; fax: +49 331 977 2092.

promising approach for groundwater remediation (Crane and Scott, 2012).

Permeable reactive barriers (PRBs) build with granular zero-valent iron (ZVI) are established technology for almost two decades (Gillham and O'Hannesin, 1994; Matheson and Tratnyek, 1994). By changing the particle size from millimetric to nanoscale, the reactivity of iron increases several orders of magnitude due to a higher surface of nZVI (Mueller and Nowack, 2010). The surface of a particle increases by a factor of 1000 by reducing particle size with the same factor, e.g. from 0.1 mm to 100 nm (Sellers et al., 2008). nZVI is highly reactive with chlorinated organic compounds (Joo and Cheng, 2006; Li et al., 2006; Wiesner and Bottero, 2007; Zhang, 2003). An advantage of direct injection of mobile colloids could be an easier installation of a PRB, since a direct injection of nZVI into a contaminated aquifer could be more cost effective and feasible than the installation of a PRB by excavation of large volumes and direct installation into the aquifer, e.g. for deep aquifers and fractured rock aquifers (Cai et al., 2006; Marcus and Bonds, 1999).

Several field applications for ZVI and nZVI have been reported to show high cleaning efficiency for different ZVI-applications, but a review concludes a need to research on nZVI technologies, since other ZVI-technologies show a higher efficiency. In that study, millimetric and micrometric iron was described to remove 97% and 91% of contamination, while nZVI applications only remove 67% of contaminants (Comba et al., 2011). Fast aggregation of nZVI to bigger aggregates and subsequent sedimentation was identified to be the main limitation for application of nZVI (Phenrat et al., 2007). Laboratory (Schrack et al., 2004), field scale experiments (Johnson et al., 2013) and field experiments (He et al., 2010; Quinn et al., 2005) show limited transport for nZVI, which might not be enough to install PRBs in-situ, since a reduction of contaminant concentrations can only be observed close to the injection well (Zhang, 2003). Different approaches have been made to enhance mobility of nZVI. One approach is to use surface modifiers, which increases electrostatic repulsion by increasing negative surface charges. Several polyanionic stabilizers are under discussion, such as carboxymethylcellulose (CMC) (He et al., 2009), guar gum (Tiraferri and Sethi, 2009), or others (Jiemvarangkul et al., 2011; Kim et al., 2009; Phenrat et al., 2008). Another approach aims for a suited carrier particle that contains nZVI, since filtration theory proposes that colloids of a size around 1 μm show the highest mobility in soils (Tufenkji and Elimelech, 2004). Detailed insight into filtration theory and nanomaterial transport is presented in reviews (Brant et al., 2007; Petosa et al., 2010). Several approaches to create a mobile composite material have been published recently: Carrier materials could consist of silica (Zheng et al., 2008), carbon black (Hoch et al., 2008), graphite (Zhang et al., 2006), carbon microspheres (Sunkara et al., 2010, 2011; Zhan et al., 2011), or activated carbon (Bleyle et al., 2012; Mackenzie et al., 2012). The last approach creates a composite material called "Carbo-Iron® colloids" (CIC), which shows material properties that are favorable for mobility. The mean particle diameter (d_{50}) of 0.8 μm (Bleyle et al., 2012) to 2.4 μm (this study) is in the relevant range for possible transport, the density is 1.7 g/cm^3 , and the composite contains 10–25 wt.% of nZVI. Additionally to that, the activated carbon is able to absorb organic compounds and to accumulate at interfaces of

groundwater and non-aqueous phase liquids (NAPL). Degradation of TCE by CIC has already been shown (Mackenzie et al., 2012).

Transport of colloids in porous media is usually described by the DLVO theory. The DLVO theory expresses the interaction energy between surfaces, e.g. particle–particle interactions or particle–surface interactions, as sum of van der Waals forces and electrostatic interactions. The DLVO theory has been extended for additional driving forces such as acid base interactions and Born repulsion forces. Additional forces, e.g. steric interactions, show additional influence on colloid transport and might be directly manipulated by macromolecules. Qualitative predictions from DLVO models suggest increased agglomeration and deposition of colloids at high ionic strength and pH values close to the point of zero charge due to reduction of repulsive forces (Brant et al., 2007). The transport and attachment of colloids in porous media is described in filtration theory by the mechanisms of interception, gravitational sedimentation and Brownian diffusion which are additive. Depending on the colloid size, the mechanisms are developed to another extent, which leads to a possible colloid size with the smallest contact and attachment probability (Tufenkji and Elimelech, 2004; Yao et al., 1971). In our study we investigate the mobility of activated carbon colloid supported nZVI (c-nZVI) in column tests using particles, produced as a variant of CIC with a bigger mean diameter of 2.4 μm than the optimal size range of 0.5–2 μm for transport in aquifer systems according to Tufenkji and Elimelech (2004). We want to examine whether an anionic stabilizer can affect the mobility of c-nZVI, if the porous media have an influence on the transport, and which environmental conditions are suited for injecting c-nZVI for groundwater remediation. Here the influence of ionic strength (NaCl and CaCl_2) and pH is examined. Additionally changes in zeta potential and particle size distribution are observed during a transport experiment.

2. Materials and methods

A sample of c-nZVI was provided by the Helmholtz Centre for Environmental Research UFZ (Leipzig, Germany). CMC, HCl, NaCl and CaCl_2 were of analytical grade (p.a.) and purchased from Carl Roth (Karlsruhe, Germany). Deionized water was produced using an ion exchange resin containing cartridge. Potassium hydrogen phthalate (KHP) was of analytical grade and provided by Elementar (Hanau, Germany).

The particle size distribution was measured by static light scattering using a Mastersizer 2000 (Malvern GmbH, Herrenberg, Germany). Suspensions were obtained by probe sonication using a Branson Sonifier 450 (Branson Ultrasonics Corporation, Danbury, CT, USA) for particle characterization. Specific surface area (BET method) was obtained by taking an ASAP 2010 accelerated surface area and porosimetry analyzer (Micromeritics GmbH, Mönchengladbach, Germany). Zeta potential was measured by using a Zetasizer Nano (Malvern GmbH, Herrenberg, Germany).

Samples for sedimentation experiments were prepared by purging 1 L of deionized water with nitrogen for 1 h to remove oxygen. For one experiment, an amount of 200 mg CMC was added and dissolved by sonication and a sample was taken for detecting background carbon concentration. A second experiment was performed without addition of CMC.

Download English Version:

<https://daneshyari.com/en/article/6386515>

Download Persian Version:

<https://daneshyari.com/article/6386515>

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