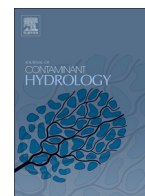




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Review article

Methods for characterizing the fate and effects of nano zerovalent iron during groundwater remediation

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ABSTRACT

The emplacement of nano zerovalent iron (nZVI) for groundwater remediation is usually monitored by common measurements such as pH, total iron content, and oxidation–reduction potential (ORP) by potentiometry. However, the interpretation of such measurements can be misleading because of the complex interactions between the target materials (e.g., suspensions of highly reactive and variably aggregated nanoparticles) and aquifer materials (sediments and groundwater), and multiple complications related to sampling and detection methods. This paper reviews current practice for both direct and indirect characterizations of nZVI during groundwater remediation and explores prospects for improving these methods and/or refining the interpretation of these measurements. To support our recommendations, results are presented based on laboratory batch and column studies of nZVI detection using chemical, electrochemical, and geophysical methods. Chemical redox probes appear to be a promising new method for specifically detecting nZVI, based on laboratory tests. The potentiometric and voltammetric detections of iron nanoparticles, using traditional stationary disc electrodes, rotating disc electrodes, and flow-through cell disc electrodes, provide insight for interpreting ORP measurements, which are affected by solution chemistry conditions and the interactions between iron nanoparticles and the electrode surface. The geophysical methods used for characterizing ZVI during groundwater remediation are reviewed and its application for nZVI detection is assessed with results of laboratory column experiments.

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1. Introduction

Nano zerovalent iron (nZVI) is one of the most extensively studied types of environmental nanoparticles because of its use in groundwater remediation. Many laboratory studies have been conducted to investigate multiple aspects related to nZVI application, including synthesis methods (He and Zhao, 2007; Wang and Zhang, 1997), material characterization of nZVI freshly prepared by various methods (Baer et al., 2010a, 2012; He and Zhao, 2005; Nurmi et al., 2005, 2011; Sarathy et al., 2008; Yan et al., 2010b), mobility in porous media (He et al., 2007, 2009; Johnson et al., 2009; Phenrat et al., 2009; Phenrat et al., 2010), and interactions/reactions with a wide range of contaminants (Fan et al., 2013, 2014; Ling and Zhang, 2014a,b; Liu and Lowry, 2006; Liu et al., 2007; Wang and Zhang, 1997). This laboratory-based work is sufficiently well-developed and abundant that it has been the basis for multiple reviews of the process-level aspects of potential applications of nZVI in remediation (O'Carroll et al., 2013; Tosco et al., 2014; Tratnyek and Johnson, 2006; Yan et al., 2013).

Compared with the advanced state of literature from process-level laboratory-scale studies with nZVI, relatively few detailed studies have been reported on the application of nZVI for groundwater remediation at the field scale. In the few studies that have been well documented (Bennett et al., 2010; Elliott and Zhang, 2001; He et al., 2010; Henn and Waddill, 2006; Johnson et al., 2013; Kocur et al., 2014; Quinn et al., 2005; Wei et al., 2010), the effectiveness of nZVI injection is usually assessed mainly by the changes in the concentrations of target contaminants of concern (CoCs), even though there are multiple processes other than degradation by nZVI that might contribute to CoC attenuation (displacement, dilution, stripping into off-gassing H₂, etc.). More comprehensive methods for characterizing the fate and effects of nZVI – including nZVI delivery, reactivity, transformation, effects on contaminants, and impact on the surrounding environment – during groundwater remediation are needed to elucidate the potential of this technology. The only study to date that emphasizes the methodological aspects of assessing nZVI transport and fate is

described in two of our previous publications (Johnson et al., 2013; Shi et al., 2011). This review is the culmination of that effort, including a synthesis of results and insights from prior work, supplemented with complementary data that we had not previously reported. It begins with a brief review of essential precursory topics (composition of nZVI formulations and reactivity of nZVI with media), followed by classification and discussion of ex situ characterization methods (performed on water samples from monitoring wells), and then in situ characterization methods (mainly geophysical). The overall goal is to advance the quality of site characterization during and after subsurface emplacements of nZVI and provide a more reliable basis for interpreting and optimizing the performance of this technology.

2. Preparation and formulation of nZVI

Most nZVI is produced with chemical reduction methods, either by H₂ under high temperature (e.g., the commercial product known as RNIP-10DP and RNIP-M2 from Toda Corp.), or by borohydride reduction of aqueous Fe(II) under ambient conditions (Fe^{BH}) (which is the most common on-site synthesis method for laboratory and field studies). Bare nZVI particles usually have a core-shell structure composed of an Fe⁰ core covered by a thin layer of iron (hydr)oxides (Li and Zhang, 2006; Martin et al., 2008; Nurmi et al., 2005). Due to electrostatic and magnetic attractions, bare nZVI particles in aqueous suspension quickly form aggregates that are large enough to have poor mobility in porous media (Phenrat et al., 2007). To minimize aggregation and maximize mobility, nZVI is typically prepared and/or deployed in the presence of additives such as surfactants or other polyelectrolytes. For example, RNIP-M2 from Toda Corp. was nZVI with surface modification by aspartate. Among numerous methods for surface modification of nZVI (reviewed in Tratnyek et al. (2011) and Yan et al. (2013)), the use of carboxymethyl cellulose (CMC) stabilizers for nZVI surface modification (CMC-nZVI) has attracted the most interest for field application (He and Zhao, 2005, 2007; He et al., 2007, 2009, 2010; Johnson et al., 2013; Kocur et al., 2014).

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