

Review

Review of soluble uranium removal by nanoscale zero valent iron

C. Jing^{a, b}, Y.L. Li^a, S. Landsberger^{b, *}^a School of Environmental Studies, China University of Geosciences, 388 Lumo Road, Wuhan, Hubei 430074, China^b Nuclear Engineering Teaching Lab, University of Texas at Austin, 10100 Burnet Road, Austin, TX 78712, USA

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ABSTRACT

Uranium (U) has been released to surface soil and groundwater through military and industrial activities. Soluble forms of U transferred to drinking water sources and food supplements can potentially threaten humans and the biosphere due to its chemical toxicity and radioactivity. The immobilization of aqueous U onto iron-based minerals is one of the most vital geochemical processes controlling the transport of U. As a consequence, much research has been focused on the use of iron-based materials for the treatment of U contaminated waters. One material currently being tested is nanoscale zero-valent iron (nZVI). However, understanding the removal mechanism of U onto nZVI is crucial to develop new technologies for contaminated water resources. This review article aims to provide information on the removal mechanism of U onto nZVI under different conditions (pH, U concentration, solution ion strength, humic acid, presence of O₂ and CO₂, microorganism effect) pertinent to environmental and engineered systems, and to provide risk or performance assessment results with the stability of nZVI products after removal of U in environmental restoration.

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1. Introduction

The nZVI has been shown to be effective for environmental remediation of a wide variety of organic and inorganic contaminants present in water resources. Some examples include toxic metals, nitrate, chlorinated organic solvents and azo-dyes. Due to

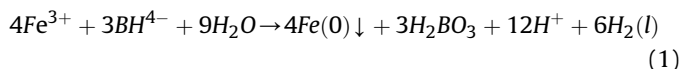
* Corresponding author.

E-mail address: s.landsberger@mail.utexas.edu (S. Landsberger).

their large surface area and high number of active sites, nZVI enhances the remediation efficiencies of those contaminants remarkably. It would appear that the first report on the use of zero valent iron (ZVI) for removal of U appeared in 1995 for remediation of contaminated groundwater. The positive result revealed that elemental iron (Fe(0)) has great potential as an inexpensive barrier material for *in situ* environmental remediation (Cantrell et al., 1995). In the next three years several other studies supported the application of ZVI to intercept U in the permeable reactive barrier technology (PRBs) (Farrell et al., 1999; Fiedor et al., 1998; Gu et al., 1998). Since 2000, ZVI has been extensively investigated to remove radionuclides from contaminated areas. Some studies demonstrated the removal effectiveness of U by ZVI from aqueous phase (Cui and Spahi, 2002; Morrison et al., 2001; Noubactep et al., 2005a, 2005b; Qiu et al., 2000; Roh et al., 2000). On the other hand, some results illustrated the gradually decreasing of reactivity on ZVI surface because of corrosion products of iron and U precipitates during contact with water. In recent years, nZVI is believed to have improved performances in restoration of U contaminated water resources. Various aspects of the removal of U on nZVI has been improved: synthesis of nZVI (Li et al., 2015; Wang and Zhang, 1997; Zhang, 2003), removal efficiency of U (Dickinson and Scott, 2010; Riba et al., 2008), kinetics and thermodynamics of removal reaction (Klimkova et al., 2011; Li et al., 2013; Yan et al., 2014), removal mechanisms (Dickinson and Scott, 2010; Klimkova et al., 2011; Li et al., 2013; Riba et al., 2008; Yan et al., 2010), influencing factors and reuse and stability of products (Crane et al., 2015a, 2015b; Li et al., 2015). While numerous laboratory and field studies have focused on the kinetics and mechanisms of the reaction between nZVI and U contaminants, few studies have included synthesized composite materials such as supported nZVI to enhance the restoration function of Fe(0) in remediation applications (Crane and Scott, 2014a; Sheng et al., 2014).

2. Synthesis of nZVI

According to previous research, there are several ways to synthesize nZVI. One method is the decomposition of iron pentacarbonyl (Fe(CO)₅) solvents into inert gas or organic agents (Choi et al., 2001; Elihn et al., 1999; Karlsson et al., 2005). Another method is the reduction of iron oxides with hydrogen. One of the most commonly used methods to prepare nZVI is the reduction of ferric iron (Fe(III)) or ferrous iron (Fe(II)) with sodium borohydride (NaBH₄) in aqueous solution via the following reaction (Li et al., 2015; Wang and Zhang, 1997; Zhang, 2003):



Using sodium borohydride to reduce Fe(III) to a metallic state is a simple way to obtain zero valent iron particles which can be safely synthesized in most chemistry laboratories with ordinary chemical reagents. It can be conducted in a flask reactor with three open necks as illustrated in Fig. 1.

The central neck is housed with a tunable mechanical stirrer in order to mix Fe(III) or Fe(II) with borohydride properly and keep the nZVI well-dispersed. Sodium borohydride drips into the reactor from one side neck (left) while inert gas is filled into the mixture from another side neck (right) to keep an anaerobic environment (Jing et al., 2015). After completion of the reaction, the products are separated from the liquid phase through a vacuum filtration and washed several times with deionized water, ethanol or acetone. The obtained black solids are vacuum dried overnight and stored under dry conditions (Dickinson and Scott, 2010; Klimkova et al., 2011; Riba et al., 2008).

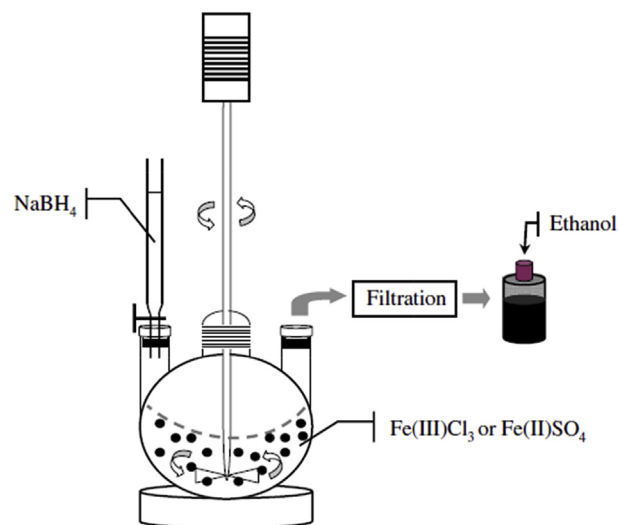


Fig. 1. Experimental setup for nZVI synthesis.

Synthesized nZVI has a core-shell construction as shown in Fig. 2. The surface of nZVI is covered with iron oxides such as FeO, Fe₂O₃ and Fe₃O₄ and its hydroxides (FeOOH) (Jing et al., 2015; Ju-Nam and Lead, 2008; Li et al., 2006a, 2006b). The Fe(0) is in the center of the particles. The relative content of products results from the mole ratio of sodium borohydride and Fe(III). At the same time, the anaerobic environment is also important to obtain more of nZVI products. Two aspects can control nZVI size and size distribution: increasing the speed of sodium borohydride addition and the speed of stirring of the reaction system. The nZVI is obtained if sodium borohydride is added carefully and the mixture stirred vigorously. Otherwise, ZVI is formed (Goldstein and Greenlee, 2012; Nurmi et al., 2005; Sun et al., 2007).

3. Removal capacity of U by nZVI

Removal capacity of U by nZVI under different conditions such as U resources, initial concentration, pH, nZVI usage, and reaction time are presented in Table 1.

Clearly U in aqueous solutions can be taken up by nZVI in a

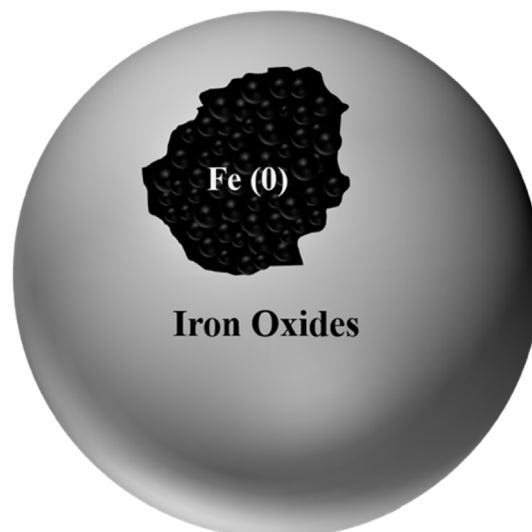


Fig. 2. The core-shell model of nZVI.

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