Environmental Research 163 (2018) 217–227

Contents lists available at ScienceDirect

Environmental Research

journal homepage: www.elsevier.com/locate/envres

Review article

Remediation of contaminated soils by enhanced nanoscale zero valent iron

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ARTICLE INFO

Keywords:
NZVI
Soil remediation
Combined pollution
Soil organisms
Enhancement

ABSTRACT

The use of nanoscale zero valent iron (nZVI) for in situ remediation of soil contamination caused by heavy metals and organic pollutants has drawn great concern, primarily owing to its potential for excellent activity, low cost and low toxicity. This reviews considers recent advances in our understanding of the role of nZVI and enhanced nZVI strategy in the remediation of heavy metals and persistent organic contaminants polluted soil. The performance, the migration and transformation of nZVI affected by the soil physical and chemical conditions are summarized. However, the addition of nZVI inevitably disturbs the soil ecosystem, thus the impacts of nZVI on soil organisms are discussed. In order to further investigate the remediation effect of nZVI, physical, chemical and biological method combination with nZVI was developed to enhance the performance of nZVI. From a high efficient and environmentally friendly perspective, biological method enhanced nZVI technology will be future research needs. Possible improvement of nZVI-based materials and potential areas for further applications in soil remediation are also proposed.

1. Introduction

In recent years, the accelerating industrialization process, the unreasonable exploitation of mineral resources and smelting emissions, long-term wastewater irrigation on soil and sludge application, dust precipitation caused by human activities, as well as the application of chemical fertilizers and pesticides, cause the soil pollution (Zhao et al., 2016; Fayiga and Saha, 2016). Soil is a main place for accumulating heavy metals and persistent organic pollutants, thus, the management and remediation of soil is critical important.

The conventional methods of treating contaminated soil include soil washing/flushing (Dermont et al., 2008; Lemaire et al., 2013), thermal desorption (Vamerali et al., 2009), vitrification (Curiel Yuste et al., 2009), photocatalyst (Wang et al., 2016, 2017; Wu et al., 2017) and bioremediation (Barnes et al., 2010), however, these methods have a risk of relatively expensive, time consuming and secondary pollution. The presence of many nanomaterials has been a boon to soil restoration (Xiong et al., 2018). However, most nanomaterials are nonmagnetic and difficult to recycle. Compared with other nanomaterials, nZVI caused extensive attention because it is non-toxic, cheap, abundant and easy to produce (Karn et al., 2011; Crane and Scott, 2012; Xu et al., 2012a; Xu et al., 2012b). Due to its nano size, nZVI has a higher reactivity towards a wide range of contaminants, especially heavy metal and organic pollutants (Fu et al., 2014; Mueller et al., 2012; Lefevre et al., 2016; Zhou et al., 2016; Guan et al., 2015; Gong et al., 2009; Xu et al., 2012a), and a higher soil mobility and delivery compared to its microscale counterpart and directly injected into the pollution source area. Consequently, nZVI is deemed as a promising remediation strategy suitable to remediate the contaminated soil. Meanwhile, organism is key contributors in soil fundamental ecosystem process. Therefore, potential issues are related to an overall evaluation of the long-term effect of nZVI on soil organism, including microorganisms (Chaithawiwat et al., 2016a, 2016b; Sacca et al., 2014; Huang et al., 2017) geobiont (El-Temsah and Joner, 2012, 2013; Yirsaw et al., 2016; El-Temsah et al., 2016) and plants (Ma et al., 2013; Monica and Cremonini, 2009; Wu et al., 2016; Li et al., 2015) have to be considered before further in situ deployment of nZVI remediation strategies. Thus, in order to reduce the excess use of nZVI influencing the adverse effect on soil organisms, researchers employed physical (Stefaniuk et al., 2016; Wang et al., 2014; Fukushima et al., 2000; Gomes et al., 2014) and chemical (Dermont et al., 2008; Lemaire et al., 2013; Danish et al., 2016; Zhang etal, 2007) methods to enhance the performance of nZVI. However, these assistive technologies needed energy consumption and caused secondary pollution, which are not favorable for field application. To address these issues, in the recent years, bio-nZVI technology referred to bioremediation coupled with the amendment of nZVI nanoparticles attract great attention for remediation in contaminated soil. One is the combined remediation strategy using phytoremediation and

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https://doi.org/10.1016/j.envres.2018.01.030
Received 20 November 2017; Received in revised form 16 January 2018; Accepted 23 January 2018

Table 1
Removal of heavy metal contaminants by nZVI in soil.

Target soil	Treatments	Pollution concentration	Main performances	Reference
As-polluted soil	nZVI	315 mg/kg	As bioaccessibility was decreased by 40.4%	Zhang et al. (2010)
As-polluted soil	nZVI	5800 mg/kg	As in the residual fraction was increased.	Gil-Diaz et al. (2016)
Pb/Zn-polluted soil	nZVI	Pb 9.0 mg/kg Zn 20 mg/kg	Residual fractions of Pb/Zn was increased	Mar Gil-Díaz et al. (2014)
Pb-polluted soil	0.2 g/L nZVI and 0.2 M citric acid	132 mg/kg	Pb efficiency removal was reached 83%.	Wang et al. (2014a)
Cr(VI)-polluted soil	nZVI/Cu	120 mg/kg	Cr(VI) reduction efficiency exceeded 99% at a pH of 5.	Zhu et al. (2016)
Cr(VI) polluted soil	0.01–0.15 g/L nZVI	15.84 mg/kg.	Cr(VI) reduction efficiency was increased from 14.51% to 86.83% in 120 min	Singh et al. (2012)
Cr-polluted soil	nZVI/Biochar	320 mg/kg,	Cr(VI) and Cr_{total} by inexpensive biochar-supported nZVI (nZVI@BC) was 100% and 91.94%	Su et al. (2016)
Cr-polluted soil	starch–stabilized nZVI	400 mg/kg.	An important reduction of Cr bioavailability	Alidokht et al. (2011)
Pb, Cd and Cr multi-polluted soil	nZVI/Activated carbon	Cd 360 mg/kg Pb 600 mg/kg Cr 80 mg/kg	Bioavailability and toxicity was reduced.	Chen et al., 2015
Sb(V)-polluted soil	nZVI and Humic acid-coated nZVI	0.685 mg/kg	The observed rate constant of was decreased in the presence of HA.	Dorjee et al. (2014)
Uranium-polluted red soil	nZVI	50 mg/kg	The adsorption capacity was increased by 5–10 times	Zhang,Liu, 2015

nZVI. Hyperaccumudated plant Panicum maximum and Helianthus annuus combined with nZVI obtained the effective remediation in TNT-contaminated soil (Jiamjitrpanich et al., 2012). In the study of Gong et al. (Gong et al., 2017b), low concentration of nZVI alleviated the oxidative damage to rumine under Cd-stress, providing the basis for combiantion phytoremediation and nZVI. Another important method is combiantion microremediation with nZVI. A rapid decrease of the pollutant concentration together with hydrogen evolution and redox potential shifts caused bt nZVI can finally lead to favorable conditions for consequent biological process (Němeček et al., 2016; Binh et al., 2016; Xiu et al., 2010). This united process could completely mineralize organic pollutants and immobilize heavy metal in a shorter time and avoid the generation of toxic byproducts, which will be the focus of future research. In this review, firstly, the performance, the migration and transformation of nZVI affected by the soil physical and chemical properties are summarized. Secondly, physically, chemically and biologically enhanced nZVI strategy are propsed. Thirdly, the toxicity of nZVI to soil organisms are discussed. Lastly, challenges and outlook of nZVI technology are well offered. With the flouring development of nZVI and enhanced nZVI strategy, their applications for soil remediation could be accessed in an effective, convenient and recyclable pathway.

2. Environment application of nZVI in soil

2.1. Removal of heavy metal contaminants by nZVI

Soil is not only an integral part of the terrestrial ecosystem but also an important reservoir of volume of pollutants. In the last few decades, soil pollution caused by heavy metals is a worldwide challenge (Wu et al., 2014). Anthropogenic activities, such as mining, military activities, manufacturing, and the long-term wastewater irrigation on soil and industrial or domestic sludge application are the main sources of metal pollution (Aminiyan et al., 2016; Fayiga and Saha, 2016; Wei and Yang, 2010; Shahid et al., 2014; Fan et al., 2008). To decrease heavy metal contaminant bioavailability and mobility, various strategies have been used (Hu et al., 2011; Huang et al., 2008; Tang et al., 2008; Feng et al., 2010). Among these, nZVI has emerged as an effective option for the treatment of contaminated soil (Zhang et al., 2010; Gil-Diaz et al., 2016; Mar Gil-Díaz et al., 2014; Fajardo et al., 2015; Chen et al., 2015). Zhang et al. (2010) found that bioaccessibility of As decreased by 40.4%, demonstrating good results for As immobility after supplement with nZVI. Similarly, the application of nZVI reduced the amount of As in the available fractions and increased the amount of As in the residual fraction (Gil-Diaz et al., 2016). The formation of adsorption complexes and precipitation process accounted for As immobilization (Jegadeesan et al., 2005). Except for As, nZVI had good immobilization effect on several common heavy metals in soil, including Pb, Zn, Se, Cr,Cd and so on (Mar Gil-Díaz et al., 2014; Mele et al., 2015; Dorjee et al., 2014; Chen et al., 2016; Su et al., 2016). Interestingly, superior immobilization effect was found for Pb than Zn under the same condition (Mar Gil-Díaz et al., 2014). The presence of soil-derived humic acid influenced the heavy metal immobilization process by nZVI due to competition for adsorption sites (Dorjee et al., 2014).

In recent year, due to the high efficiency, bimetal nZVI nanoparticles used for heavy metal soil remediation has brought about widespread attention. Evidence, to date, suggested as the contaminant concentration are high up to 120 mg/kg, Cr(VI) reduction efficiency exceeded 99% under weakly acidic conditions as the nZVI/Cu nanoparticles was used in the contaminated soil (Zhu et al., 2016). nZVI, as a powerful, inexpensively and environmental friendly agent, has been used for heavy metal soil remediation, and satisfactory results have been achieved not only in the laboratory but also in the field. Singh conducted the success field application for the reduction of Cr(VI) in contaminated soil with nZVI and obtained satisfactory results (Singh et al., 2012). Zhang confirmed the feasibility of nZVI as the reactive barriers (PBR) from in situ remediation of uranium-contaminated red soils. The removal capacity of U(VI) polluted soil with supplement with nZVI was significantly higher (5–10 times) than those of soil without supplement (Zhang et al., 2015). Unfortunately, the technology limitations which include poor stability and mobility of nZVI and tendency of aggregation, further reduced the reduction reactivity in soil. In conclusion, the use of nZVI to remediate polluted soils with heavy metals is a promising in situ strategy. However, additional work is required to enhance the performance of nZVI.(Tables 1, 2)

2.2. Removal of persistent organic pollutants (POPs) by nZVI

Besides heavy metals, persistent organic pollutants also pose a serious threat to the human health and environment safety, especially in soil (Solé et al., 2013; Jwg et al., 2011; Zeng et al., 2013b; Bokare et al., 2010).Owing to these compounds of extreme persistence and recalcitrance, there is urgent to develop a cost effective and sustainable remediation technology, and nZVI is considered to be a promising alternative for reductive degradation of POPs.

Recalcitrant polycyclic aromatic hydrocarbons (PAHs) such as Benzoapyrene (BaP) and Anthracene (ANT) were completely depleted with persulfate activated by nZVI, however, Phenanthrene (PHE)