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Review

Application of iron sulfide particles for groundwater and soil remediation: A review

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ABSTRACT

Rapid industrialization and urbanization have resulted in elevated concentrations of hazardous inorganic and organic contaminants in groundwater and soil, which has become a paramount concern to the environment and the public health. In recent years, iron sulfide (FeS), a major constituent of acid-volatile sulfides, has elicited extensive interests in environmental remediation due to its ubiquitous presence and high treatment efficiency in anoxic environment. This paper provides a comprehensive review on recent advances in: (1) synthesis of FeS particles (including nanoscale FeS); and (2) reactivity of FeS towards a variety of common environmental contaminants in groundwater and soil over extended periods of time, namely, heavy metals (Hg(II), Cu(II), Pb(II), and Cr(VI)), oxyanions (arsenite, arsenate, selenite, and selenate), radionuclides (e.g., uranium (U) and neptunium (Np)), chlorinated organic compounds (e.g., trichloroethane, trichloroethylene, and *p*-chloroaniline), nitroaromatic compounds, and polychlorinated biphenyls. Different physiochemical and biological methods for preparing FeS with desired particle size, structure, and surface properties are discussed. Reaction principles and removal effectiveness/constraints are discussed in details. Special attention is placed to the application of nanoscale FeS particles because of their unique properties, such as small particle size, large specific surface area, high surface reactivity, and soil deliverability in the subsurface. Moreover, current knowledge gaps and further research needs are identified.

Contents

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List of acronyms

CMC	Carboxymethyl cellulose
COCs	Chlorinated organic compounds
CT	Carbon tetrachloride
DNT	2,4-dinitrotoluene
EPA	Environmental Protection Agency
HA	Humic acid
HCA	Hexachloroethane
IS	Ionic strength
MCL	Maximum contaminant level
MeHg	Methylmercury
NACs	Nitroaromatic compounds
PCA	*p*-chloroaniline
PCBs	Polychlorinated biphenyls
PCE	Tetrachloroethylene
PRB	Permeable reactive barriers
SEM	Scanning electron microscope
SRB	Sulfate reducing bacteria
TCA	Trichloroethane
TCE	Trichloroethylene
TCLP	Toxicity characteristic leaching procedure
TEM	Transmission electron microscopy
TOC	Total organic carbon
ZVI	Zero-valent iron

1. Introduction

Soil and groundwater contamination has become a major environmental concern because of the enormous scale and the complex environmental conditions. In USA, it was estimated that there are 350,000 contaminated sites requiring cleanup over the next 30 years (USEPA, 2012). In Europe, there are 3,000,000 potential contaminated sites, of which >8% (~250,000 sites) are highly contaminated and need remediation (EEA, 2007). Iron sulfide (FeS) or mackinawite, a tetragonal ferrous monosulfide, is the first crystalline ferrous sulfide solid phase to form under sulfate reducing conditions (Jeong et al., 2008; Rickard, 1995). FeS is a ubiquitous, non-toxic mineral and a precursor to other stable iron sulfide minerals, such as pyrite and greigite. Moreover, FeS can be easily synthesized. Consequently, FeS has been applied for treatment of groundwater and soil contaminated with heavy metals, chlorinated organic compounds (COCs), arsenic, selenium, and other inorganic and organic contaminants (Table 1).

Table 1
Examples of common contaminants in groundwater and soil remediated by FeS.

Groups	Pollutants	Removal mechanisms	References
Inorganic compounds			
Heavy metals	Mercury (Hg)	Chemical precipitation: $FeS(s)+Hg^{2+}\Leftrightarrow HgS(s)+Fe^{2+}$ Ion exchange: $FeS(s)+xHg^{2+}\Leftrightarrow [Fe_{1-x},Hg_x]S(s)+xFe^{2+}$ (x<1) Surface complexation: $FeS(s)+Hg^{2+}\Leftrightarrow FeS-Hg^{2+}$	(Jeong and Hayes, 2007; Skyllberg and Drott, 2010)
	Chromium (Cr)	Reduction: $FeS(s)+Cr^{6+}\Leftrightarrow S+Fe^{3+}+Cr^{3+}$ Chemical precipitation (pH>4): $xCr^{3+}+(1-x)Fe^{3+}+3H_2O\Leftrightarrow (Cr_xFe_{1-x})(OH)_3(s)+3H^+$ (x<1)	(Mullet et al., 2004)
Inorganic oxyanions	Arsenic (As)		
	As(III)	Chemical precipitation (pH=5):$3FeS+H_3AsO_3+3H^+\Leftrightarrow 1/2Fe_3S_4+AsS+3/2Fe^{2+}+3H_2O$ Surface sorption (pH>6)	(Han et al., 2011a)
	As(V)	Outer-sphere complexation and chemical precipitation	(Wolthers et al., 2005)
	Selenium (Se)		
	Se(IV)	Reduction and chemical precipitation	(Breynaert et al., 2008; Han et al., 2011b; Scheinost and Charlet, 2008)
	Se(VI)	Surface sorption	(Han et al., 2011b)
Radionuclides	Uranium (U)	Reduction: $UO_2^{2+}+\equiv FeS\Leftrightarrow\equiv S^{2-}-UO_2^{2+}+Fe^{2+}\equiv S^{2-}-UO_2^{2+}\Leftrightarrow S^0(s)-UO_2(s)$ $FeS(s)+H_2O\Leftrightarrow Fe^{2+}+HS^-+OH^-\ UO_2^{2+}+HS^-\Leftrightarrow UO_2(s)-S^0(s)+H^+$	(Hyun et al., 2012)
	Technetium (Tc)	Reduction and chemical precipitation	(Livens et al., 2004)
	Neptunium (Np)	Reduction and surface complexation	(Livens et al., 2004; Moyes et al., 2000)
Organic compounds			
Chlorinated organic compounds (COCs)	Trichloroethylene (TCE)	Reduction	(Butler and Hayes, 1999; Jeong et al., 2007a)
	Tetrachloroethylene (PCE)	Reduction	(Butler and Hayes, 1999; Jeong et al., 2007a)
Nitroaromatic compounds (NACs)		Reduction	(Oh et al., 2011)
Polychlorinated biphenyls (PCBs)		Reduction	(Watson et al., 2002)