



Remediation of hexavalent chromium spiked soil by using synthesized iron sulfide particles



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HIGHLIGHTS

- CMC stabilized FeS particles are synthesized and characterized.
- The effects of remediation process factors are discussed.
- Synthesized FeS particles offer an effective immobilization of Cr(VI) in soil.
- FeS particles and FeSO₄ are compared for Cr(VI) contaminated soil remediation.
- Synthesized FeS particles cause less pH change and more stable Cr fractions in soil.

ARTICLE INFO

Article history:

Received 6 April 2016

Received in revised form

6 November 2016

Accepted 11 November 2016

Available online 18 November 2016

Handling Editor: X. Cao

Keywords:

Hexavalent chromium/Cr(VI)

FeS particles

Soil remediation

Immobilization

ABSTRACT

Carboxymethyl cellulose (CMC) stabilized microscale iron sulfide (FeS) particles were synthesized and applied to remediate hexavalent chromium (Cr(VI)) spiked soil. The effects of parameters including dosage of FeS particles, soil moisture, and natural organic matter (NOM) in soil were investigated with comparison to iron sulfate (FeSO₄). The results show that the stabilized FeS particles can reduce Cr(VI) and immobilize Cr in soil quickly and efficiently. The soil moisture ranging from 40% to 70% and NOM in soil had no significant effects on Cr(VI) remediation by FeS particles. When molar ratio of FeS to Cr(VI) was 1.5:1, about 98% of Cr(VI) in soil was reduced by FeS particles in 3 d and Cr(VI) concentration decreased from 1407 mg kg⁻¹ to 16 mg kg⁻¹. The total Cr and Cr(VI) in Toxicity Characteristic Leaching Procedure (TCLP) leachate were reduced by 98.4% and 99.4%, respectively. In FeS particles-treated soil, the exchangeable Cr fraction was mainly converted to Fe-Mn oxides bound fraction because of the precipitation of Cr(III)-Fe(III) hydroxides. The physiologically based extraction test (PBET) bioaccessibility of Cr was decreased from 58.67% to 6.98%. Compared to FeSO₄, the high Cr(VI) removal and Cr immobilization efficiency makes prepared FeS particles a great potential in field application of Cr(VI) contaminated soil remediation.

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1. Introduction

Chromium (Cr) is one of the most common heavy metals used in many industrial facilities, such as wood preservation, chrome plating and alloy formation (Fruchter, 2002). The uncontrolled treatment of Chromite ore processing residue and Cr-containing wastewater from chromium producing or consuming industries can lead to the Cr contamination of the surrounding soil (Farmer

et al., 2002; Saha and Orvig, 2010). The contamination of Cr causes the accumulation of chromium in plants and has captured a worldwide attention for its extensive scope (Fruchter, 2002; Barrera-Díaz et al., 2012). Via plants, it enters the food chain and presents potential threat to human's health. Cr is considered to be one of the top 20 contaminants on the Superfund priority list of hazardous substances for the past 15 years (Dhal et al., 2013).

Hexavalent Cr(VI) and trivalent Cr(III) are the two oxidation states of Cr in natural environment, with distinct bioavailability and toxicity (Choppala et al., 2013). Cr(VI) is highly toxic, mutagenic and teratogenic, and it primarily exists as HCrO₄⁻ at acidic pH and CrO₄²⁻

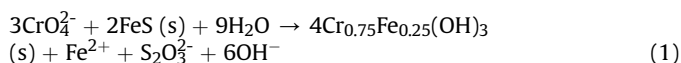
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at neutral and alkaline pH (Rai et al., 1989). These anions are hard to be adsorbed to soil colloids with negative charges. Cr(III) is known to have 100-fold lower toxicity than Cr(VI) due to its limited water solubility and mobility. In common environmental conditions (pH 6–9), Cr(III) is normally found in the form of Cr(OH)₃, which is also of relatively low solubility ($k_{sp} = 6.7 \times 10^{-31}$) and generally considered amorphous. It binds with ligands such as acetic and tartaric acid, and forms relatively stable complex with low available for plant uptake (Leita et al., 2011; Taghipour and Jalali, 2016). As Cr(III) is the species of Cr with low mobility and toxicity compared to other ones, reduction of Cr(VI) to Cr(III) is used as the main mechanism to remediate Cr(VI) contaminated soil.

Chemical reduction is the most commonly used approach for the remediation of Cr(VI) contaminated sites, by which inorganic or organic electron donors reduce Cr(VI) to Cr(III), and insoluble Cr(III) hydroxides are formed. In general, there are three groups of reducing agents for the transformation of Cr(VI) to Cr(III) including organic compounds such as ascorbate, soil organic matter and composts (Bolan et al., 2003; Xu et al., 2004; Zhang et al., 2012; Scaglia et al., 2013), reduced sulfur compounds such as sodium sulfide (Na₂S) (Lan et al., 2005) and calcium polysulfide (CaS_x) (Chrysochoou et al., 2010; Chrysochoou and Johnston, 2015), and iron-based materials such as zero-valent iron nanoparticles (nZVI) (Xu and Zhao, 2007; Singh et al., 2011; Du et al., 2012; Wang et al., 2014a,b) and dissolved ferrous iron (Dermatas et al., 2006; Moon et al., 2009; Di Palma et al., 2015) and solids containing ferrous iron (Jung et al., 2007; Mullet et al., 2007; Liu et al., 2015). The sulfur compounds reduce Cr(VI) to Cr(III), which usually form Cr(III) hydroxides with lower mobility and toxicity (Zhou et al., 2012). Besides, the iron-based materials may efficiently reduce Cr(VI) to Cr(III) and form a passive layer of Cr_xFe_{1-x}(OH)₃ or Cr_xFe_{1-x}OOH with ferric iron (Fe(III)) at the surface of solid products (Mullet et al., 2007; Di Palma et al., 2015). The Cr(III)-Fe(III) hydroxides are of extended stability in the pH range from 4.8 to 13.5, more stable than amorphous Cr(III) hydroxides (Papassiopi et al., 2014). nZVI is seen as an effective reductant for the immobilization of Cr(VI), however it is usually prepared by a relatively expensive method, borohydride reduction (ReyhaniTabara et al., 2012), and it may have a harmful effect on microorganisms, animal cells, plant cells, and human cells (Stefaniuk et al., 2016).

Iron sulfide minerals, such as pyrite (FeS₂) (Mullet et al., 2007; Liu et al., 2015) and mackinawite (FeS_{1-x}) (Boursiquot et al., 2002; Mullet et al., 2004), have been widely applied to reduce Cr(VI) in aqueous solution. FeS as a common iron-based material may act to immobilize Cr(VI) in environment with a relatively high reducing capacity. Patterson et al. (1997) indicated that amorphous FeS effectively removed Cr(VI) in solution at initial Cr(VI) concentrations ranging from 50 to 5000 μM at pH from 5 to 8 by the mechanism that Cr(VI) was reduced and precipitated by FeS following the reaction in Equation (1):



Carboxymethyl cellulose (CMC) is a kind of anionic high-polymer cellulose and frequently applied as stabilizer in application suspensions, because numerous carboxyls and hydroxyls exist in the macromolecular chain of CMC, which can coordinate with multivalent cations, such as Fe³⁺ and Al³⁺ (Gillies et al., 2016; Ren et al., 2016). Compared to FeS minerals, the diminutive size and large specific surface area of FeS particles can contribute to their enhanced reactivity (Rivero-Hugue and Marshall, 2009). Compared to soluble reducing agents such as ferrous and sulfide, FeS particles may not only gradually release the ferrous and sulfide ions to reduce Cr(VI), but also sorb Cr(VI) through surface reaction (Han

et al., 2014). FeS has been frequently applied in Cr(VI) removal from aqueous solutions, however limited studies on the utilization of FeS particles in remediation of Cr(VI) contaminated soil were reported.

The overall goal of this study was to examine the effectiveness of the remediation process using FeS particles with a micro-level size to immobilize Cr(VI) in soil and explore the effects of the remediation conditions. The specific objectives were as follows: (1) prepare and characterize CMC stabilized FeS particles in lab; (2) examine the effects of FeS particles dosage, moisture and NOM in soil on Cr(VI) removal; (3) better understand the remediation mechanisms of Cr(VI); and (4) assess the efficiency of FeS compared to FeSO₄ in the remediation of Cr(VI) spiked soil.

2. Materials and methods

2.1. Materials

All reagents except for CMC were of analytical or higher grade. The sodium form of CMC (M.W. = 90,000) was chemical grade and purchased from Qiangshun Chemical (Shanghai, China). All solutions were prepared with deionized water (18.25 MΩ cm⁻¹).

The Cr(VI) spiked soil was prepared by adding potassium dichromate to raw soil samples following the procedure of Wang et al. (2014a). The concentrations of Cr(VI) and total Cr in Cr(VI) spiked soil were 1407 mg kg⁻¹ and 2089 mg kg⁻¹, respectively. The raw soil samples were collected from a farm in Chongqing, China, SP (29°13'19.08" N and 106°15'52.85" E, altitude 409 m). The farm is used for agriculture and no industrial use was involved in history. Before use, the soil samples were air-dried and sieved with a 2-mm standard mesh, and then characterized for physicochemical properties. The pH of raw soil was 6.75 and the cation exchange capacity was 18.8 mmol kg⁻¹. The raw soil contains 0.63% of organic carbon and was initially free of Cr(VI) and Hg. The contents of Cr, Pb and Cd in raw soil were 63 mg kg⁻¹, 71 mg kg⁻¹ and 5.3 mg kg⁻¹, respectively. The soil was silty loam with 22.2 wt % sand, 60.5 wt % silt and 17.3 wt % clay.

2.2. Synthesis and characterization of FeS particles

FeS particles were prepared following the procedures adapted from the method of Xiong et al. (2009) and Gong et al. (2012). FeSO₄ solution (20 mL, 0.852 M) was mixed with deoxygenated CMC solution (110 mL, 0.2%, w/w). The mixture was then purged with nitrogen gas (99.99% pure) for 30 min to complete the formation of Fe²⁺-CMC complexes. Then Na₂S solution (20 mL, 0.852 M) was titrated into the Fe²⁺-CMC solution at a speed of 0.05 mL s⁻¹ to yield the FeS particles. The FeS particles-CMC suspension was centrifuged at 1000 rpm, and the supernatant was decanted and CMC solution (1%, w/w) was added to keep a constant volume. The mixture was sonicated for half an hour to restabilize the FeS particles with ultrasonic instrument (KQ-200KDE, Kunshan Ultrasonic Instrument, China).

The FeS particle solids were collected by filtering the suspension using a 0.45 μm membrane filter and subsequently freeze-dried under vacuum at -50 °C using a freeze dryer (MudoIYOD-230, Thermo Fisher, American) for 48 h. The X-ray diffraction (XRD) patterns of FeS particles and CMC were obtained on a Shimadzu XRD-7000 device at a scan speed of 2.0° min⁻¹. The XRD patterns were processed using the computer program known as MDI Jade 6.5 loaded with ICDD database (Materials Data Inc., Livermore, CA, U.S.A.). The surface composition and morphology were investigated by scanning electron microscopy (SEM) and energy dispersive X-ray (EDX) analysis at 15 kV (JSM-7800F, JEOL, Japan). The mean hydrodynamic diameter of FeS particles were determined by Nano

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