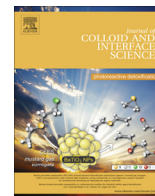




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Enhanced reduction and adsorption of hexavalent chromium by palladium and silicon rich biochar supported nanoscale zero-valent iron

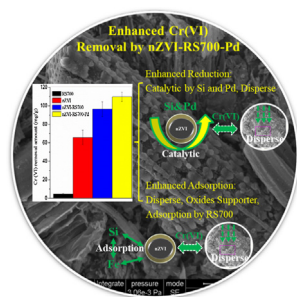


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GRAPHICAL ABSTRACT

The mechanism of Cr(VI) removal enhanced by Pb and for silicon rich biochar supported nanoscale zero-valent iron.



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ABSTRACT

The potential of silicon-rich biochar and Pd were evaluated for the enhanced removal of Cr(VI) in solution by nanoscale zero-valent iron (nZVI) composites. The composition and structures of the nZVI, RS700-supported nZVI, and Pd-doped samples were analyzed by scanning electron microscopy, Fourier-transform infrared spectroscopy, X-ray diffraction, and X-ray photoelectron spectroscopy before and after reaction with Cr(VI). The amount of Cr(VI) removed by nZVI-RS700-Pd was considerably greater than the removal by nZVI, nZVI-Pd, or nZVI-RS700. This was mainly due to the enhanced reduction and adsorption of Cr(VI) by silicon-rich biochar and Pd. Silicon and Pd promoted the reduction of Cr(VI) due to the Fe⁰ crystallinity in the nZVI structures. The significantly decreased removal of Cr(VI) by the silicon-removed sample (nZVI-RS700 (-Si)) further confirmed that silicon played a significant role in the removal of Cr(VI). Cr(VI) adsorption was enhanced by the dispersion and adsorption of RS700. Following the reaction of RS700-supported nZVI with Cr(VI), ferrous chromite (FeCr₂O₄) was observed on the nZVI-RS700 composite surface. The formation of FeCr₂O₄ can be attributed to the reduction of Cr(VI) by the nZVI and coprecipitation of chromium oxide with iron on the RS700 surface. Therefore, nZVI-RS700-Pd is a potential remediation reagent that can be used to effectively treat Cr(VI)-contaminated groundwater.

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1. Introduction

Engineered nanoscale zero-valent iron (nZVI) materials have received significant attention for their high activity and low

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environmental impact [1–4]. These nZVI composites can be used to remove a variety of pollutants via chemical reduction [5–11]. However, their rapid loss of reactivity due to aggregation severely limits applications in groundwater remediation [12,13]. Previous studies showed that the activity of nZVI can be improved using biochar, which promotes the removal of heavy metals and organic pollutants [14–16].

Many biochars, such as those derived from rice straws, contain a number of silicon minerals [17–19]. It has been reported that the silicon in biochar may serve as support sites for nZVI [16,20–24]. Generally, the reactivity of nZVI can be promoted by combination with Pd, Cu, or Ni to form bimetallic nanoparticles [22,23]. Of these metal additives, Pd-doped nZVI exhibits higher reactivity than other bimetallic systems. Pd acts as a catalyst for hydrogen formation, which reduces the charge during hydrogenation of the pollutants [24]. However, few published reports are available that describe the influence of silicon in biochar on nZVI activity or the mechanism of Cr(VI) removal. In addition, the effect of Pd on the enhancement of Cr(VI) removal by biochar-supported nZVI composites warrants further investigation.

The objective of this study was to elucidate the mechanism underlying the enhanced Cr(VI) removal by Pd modified silicon rich biochar-supported nZVI. The silicon rich biochar was derived from rice straw (pyrolyzed at 700 °C, referred as RS700). Cr(VI) removal by nZVI, RS700-supported nZVI, and Pd doped samples were investigated under a variety of pH conditions and Cr(VI) concentrations. The surface features of the samples were subsequently analyzed before and after interaction with Cr(VI). Finally, a potential mechanism was proposed for the enhanced Cr(VI) removal by nZVI, silicon, and Pd.

2. Materials and methods

2.1. Sample preparations

Biochars were produced using a previously reported method [16,25]. The rice straws were air-dried and ground before being passed through a 0.154-mm sieve. The rice straw was pyrolyzed at 700 °C under oxygen limiting conditions. The biochar produced at 700 °C was labeled as RS700 and 2 g of RS700 was then treated in 50 mL HCl (1.0 mol/L) to remove the mineral constituents and increase the surface area. The supernatant was then removed by centrifugation for 4 h. To remove insoluble silicon oxides, the RS700 after acid treatment was transferred into 25 mL of HF (1.0 mol/L) and the supernatant was removed by centrifugation after 24 h. Then the RS700 treated both in HCl and HF was washed to remove soluble salts, residual acids, and silicon. After several washes, a stable pH was obtained. The samples were subsequently oven-dried at 80 °C then passed through a 0.154-mm sieve and classified as RS700 and RS700 (-Si), with the latter treated with HF to remove silicon.

The synthesis of RS700 and RS700 (-Si) supported nZVI was performed according to the methods described in a previous study [16]. First, 2 g of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ was added to 50 mL of distilled water. After the complete dissolution, 0.1 g of RS700 or RS700 (-Si) was added to the solution. The solution pH was adjusted to 4.0 and the beaker was placed in a shaker at 150 rpm with the temperature adjusted to 25 °C. After 24 h of agitation, 50 mL of ethanol was added. All solutions were then transferred to 500 mL three neck flasks. To remove dissolved O_2 , N_2 was flushed through the solution for more than 1 h under vigorous stirring. Next, a 100 mL KBH_4 solution (0.5 mol/L, approximately 5 mL/min) was added dropwise under intense stirring to reduce ferrous iron to zero-valent iron. After the solution was added, stirring was maintained for 0.5 h. Subsequently, the solution and solid were separated using a mag-

net, the solid was washed three times and vacuum dried. The samples are hereafter referred to as nZVI-RS700 and nZVI-RS700 (-Si). In addition, nZVI without biochar support was also synthesized.

Pd-loaded nZVI and nZVI-RS700 were synthesized in the following steps. Based on the preparation of nZVI and nZVI-RS700, the Pd (approximately 1.5% of the iron content) was added to the newly prepared samples. After 10 min of ultrasound, the supernatant was removed and vacuum dried. Pd-doped nZVI and nZVI-RS700 are hereafter referred to as nZVI-Pd and nZVI-RS700-Pd.

2.2. Sample characterization

The nZVI, RS700-supported nZVI, and the Pd doped samples were characterized by Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD), X-ray photoelectron spectra (XPS), transmission electron microscopy (TEM), and scanning electron microscopy (SEM). After Cr(VI)-loading, the samples were characterized again. For analysis of the FTIR spectra (Thermo Scientific 7600, USA), KBr was used as supporter where the mass ratio of the sample to KBr was approximately 1%. To analyze the crystal structure of the samples, XRD (RIGAKU D/MAX 2550/PC, Japan) images were taken from 2 to 60°. The chemical bonding state at the sample surface was determined via XPS (Shimadzu AXIS UltraDL, Japan) with Mg K α radiation emitted from a double anode at 50 W. A full survey of the samples was obtained and analyzed at the Fe 2p, Cr 2p, Si 2p, Pd 3d, and O 1s regions. SEM-EDS (QUANTA 200, Netherlands) was used to examine the morphology and composition (including the C, Si, Fe, Cr, and O content) of the samples. TEM (Hitachi HT7700, Japan) was used to determine the iron particle sizes and morphology at an operating voltage of 100 kV. Furthermore, the soluble silicon of RS700 was determined by ICP-MS after digestion in HNO_3 at 150 °C. The content of soluble silicon in the RS700 was determined to be 4.31%. The Pd concentration was also determined by ICP-MS after digestion in $\text{HCl}/\text{HNO}_3 = 3/1$ at 180 °C. The Pd content in nZVI-Pd and nZVI-RS700-Pd was 1.26 and 1.16%, respectively.

2.3. Batch removal experiments

2.3.1. Concentration-dependent removal experiments

The effects of different Cr(VI) concentrations on the removal of chromium by nZVI, nZVI-RS700, and the Pd-doped samples were performed in 20 mL vials. The initial concentrations of Cr(VI) ($\text{K}_2\text{Cr}_2\text{O}_7$) were 20, 30, 40, 50, 60, 80, and 100 mg/L. The ratio of the sample to solution was 1:5, and the composition of the groundwater was simulated by the addition of 0.01 mol/L NaCl. Before the addition of the samples, the initial pH was adjusted to 3.5. Subsequently, the samples were added to the solution and the vials were agitated at 150 rpm using a shaker. The temperature of the shaker was maintained at 25 °C. Previous studies have shown that the equilibrium time was less than 8 h [16,26]. After 8 h, the solution was filtered using a microporous filter. The Cr(VI) concentration was determined by diphenyl-carbazide colorimetry at a detection wavelength was 540 nm. The pH of the filtrate was also determined.

2.3.2. Effect of pH

Cr(VI) removal by nZVI, RS700-supported nZVI, and Pd doped samples was investigated in 20 mL vials. The initial pH range was adjusted to 3.0–4.5, as the Cr(VI) removal reaches a minimum above pH 4.5, and the initial Cr(VI) concentration was 50 mg/L. Subsequently, the solid to liquid ratio, the concentration of the background solution, and the treatment after the sample addition were identical to the concentration-dependent removal experiments.

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