



Decontamination of U(VI) from nZVI/CNF composites investigated by batch, spectroscopic and modeling techniques

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ABSTRACT

The nZVI/CNF composites were synthesized by borohydride reduction of ferrous salts in the CNF suspension. The characteristic results indicated that metallic Fe⁰ was uniformly dispersed the surface of CNFs. The decontamination mechanism of U(VI) from nZVI/CNF composites was investigated by batch, XPS, EXAFS and modeling techniques. The batch experiments indicated that removal amount of U(VI) on nZVI/CNF was significantly higher than that of single nZVI, whereas reduction amount of U(VI) to U(IV) on nZVI at pH > 6.0 was significantly higher than that of nZVI/CNF. The EXAFS spectra of U-nZVI at pH 7.0 was similar to the U^(IV)O₂(s), whereas the EXAFS spectra of U-nZVI/CNF at 2.98 Å can be satisfactorily fitted by U-U shell, which was further demonstrated by XANES analysis and surface complexation modeling. The diffuse layer model gave better fits to U(VI) removal on nZVI/CNFs composites, whereas underestimation of U(VI) removal on nZVI was observed at pH > 5.0 due to the presence of reductive precipitation. These findings indicated that nZVI-based composites presented the higher adsorption performance and the excellent reduction capacity for U(VI) under permeable reactive barrier system.

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1. Introduction

With the rapid industrialization and urbanization of human have resulted in the contamination of radioactive pollutants into groundwater and soil, which could be seriously dangerous to the biological diversity and human health such as toxicity and cancer risks [1]. Uranium as a typical radioactive element is ubiquitous at uranium mining, processing and milling sites. Generally, uranium exhibited two oxidative state, including dissolved hexavalent uranyl (UO₂²⁺) and sparingly soluble tetravalent reductive precipitation (e.g., UO₂(s)). Therefore, the reduction of U(VI) to U(IV) is a significant pathway for the removal of uranium from aqueous solutions in environmental remediation strategy. It is demonstrated that the bio-reduction of U(VI) to U(IV) was effective way to immobilize the uranium [2–5]. However, the U(IV) was rapidly re-oxidized to U(VI) when dissolved oxygen (~5.0 mg/L) entered the injection well [3].

In recent years, nanoscale zero-valent iron (nZVI), a major constituent of nano-materials in permeable reactive barrier (PRB) system, has attracted much more attention for its potential applications in environmental remediation due to large surface area, strong redox capacity and high treated efficiency in aquatic geological repository. It has been

proven that nZVI as highly effective nanoparticles for the removal/degradation of a wide range of environmental contaminants such organic pollutants [6–10], heavy metals [11–14] and radionuclides [15–18]. Generally, nZVI nanoparticle displays core-shell structure of a metallic iron core encapsulated by an iron oxide shell [19–21]. The metallic Fe⁰ as electron donor offers a reductive capacity, whereas the iron oxide shell inhibits the electron transfer from the Fe⁰ core to the outside shell [22–24]. In additions, the nZVI nanoparticles were easily agglomerated each other, which permitted the solutes adsorption through electrostatic interaction and/or surface complexation [25,26]. To inhibit the further surface oxidation and to enhance its dispersibility in aqueous solutions, as significant advance have been achieved in the synthesis of nZVI-based composites to enhance the capacity and longevity such as nZVI/C [27], nZVI/graphene oxide composites [25], and nZVI/CNF composites [28]. However, few studies regarding the interaction mechanism of U(VI) on nZVI/CNF composites using XANES, EXAFS and modeling were available.

The objectives of this study were (1) to synthesize the nZVI/CNF composites and characterize them by XRD, SEM, TEM, and FT-IR techniques; (2) to elucidate the effect of water chemistry (reaction time, pH, ionic strength, temperature) on U(VI) interaction with nZVI and nZVI/CNF composite using batch techniques; (3) to determine the interaction mechanism of U(VI) and nZVI/CNF composites by XPS, XANES and EXAFS technique; and (4) to simulate the interaction of U(VI) on

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nZVI/CNF composites by surface complexation modeling. The highlight of this study is to distinguish the reduction from adsorption by using advanced spectroscopic and modeling techniques. This paper provides a comprehensive understanding of interaction mechanism of nZVI/CNF composites and U(VI) at water-solid interface, which are crucial for the potential application of nZVI-based nanomaterials as the high effective adsorbents for removal of radionuclides in environmental cleanup.

2. Materials and methods

2.1. Materials

Bacterial cellulose (BC, as precursor of CNF) was obtained from Hainan Yeguo Food Co., Ltd. $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ and NaBH_4 (analytical reagents) were purchased from Sinopharm Chemical Reagent Co., Ltd. The U(VI) stock solution (0.1 mmol/L) was prepared under an anaerobic conditions by dissolving the $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (99.99% purity, Sigma-Aldrich, USA) into 0.01 mol/L HNO_3 solution. Other chemicals were commercially purchased as analytical reagents from Sinopharm Chemical Reagent Co., Ltd. and used as received with further purification.

2.2. Synthesis of nZVI/CNF composites

nZVI/CNF composites were synthesized according to the modified method reported by Wang and Zhang [6]. Firstly of all, the CNF was obtained by the pyrolysis of BC pellicles under N_2 atmosphere [29,30]. Typically, the white bacterial cellulose pellicles were rinsed several times by Milli-Q water, and then bacterial cellulose pellicles were heated to 773 K

for 1 h under a flowing N_2 atmosphere. The CNFs was obtained by washing it with 1.0 mol/L HCl and then cooling it until to room temperature naturally. nZVI nanoparticles were synthesized by reduction of Fe(III) to Fe^0 by using NaBH_4 under the N_2 conditions. Typically, the NaBH_4 was slowly added into the 1.0 mol/L FeCl_3 solution under N_2 atmosphere and vigorous stirring conditions. Then the redundant salts were washed by N_2 purged Milli-Q water several times and was dried it in the vacuum oven. The nZVI/CNF composites were synthesized by NaBH_4 reduction of FeCl_3 in the CNF suspensions. Briefly, 1.0 mol/L FeCl_3 solution was dissolved into CNF suspensions, and then NaBH_4 was slowly added into aforementioned solutions under N_2 conditions. The nZVI/CNF composites were obtained by centrifugation and then was dried in vacuum oven overnight. For the comparison, the nZVI nanoparticles were also synthesized by previous studies.

2.3. Characterization

The morphology of the nZVI/CNF composites were characterized by SEM (FEI-JSM 6320 F) and TEM (Hitachi H7650 transmission electron microscope). The samples of TEM observation were prepared by suspending samples on ethanol and adding a drop of suspension onto Cu grids with holey carbon films. The identification of nZVI/CNF composites was recorded by Rigaku X-ray diffractometer with monochromatized $\text{Cu-K}\alpha$ radiation at 40 mV and 45 mA with a step size of 0.02° and a count time of 5 s per step. The types of oxygen-containing functional groups of nZVI/CNF composites were recorded by FT-IR (Nicolet 8700 FT-IR spectrometer) in pressed KBr pellets (Aldrich, 99%, FT-IR grade). Briefly, 2.0 mg of sample and 200 mg of KBr were

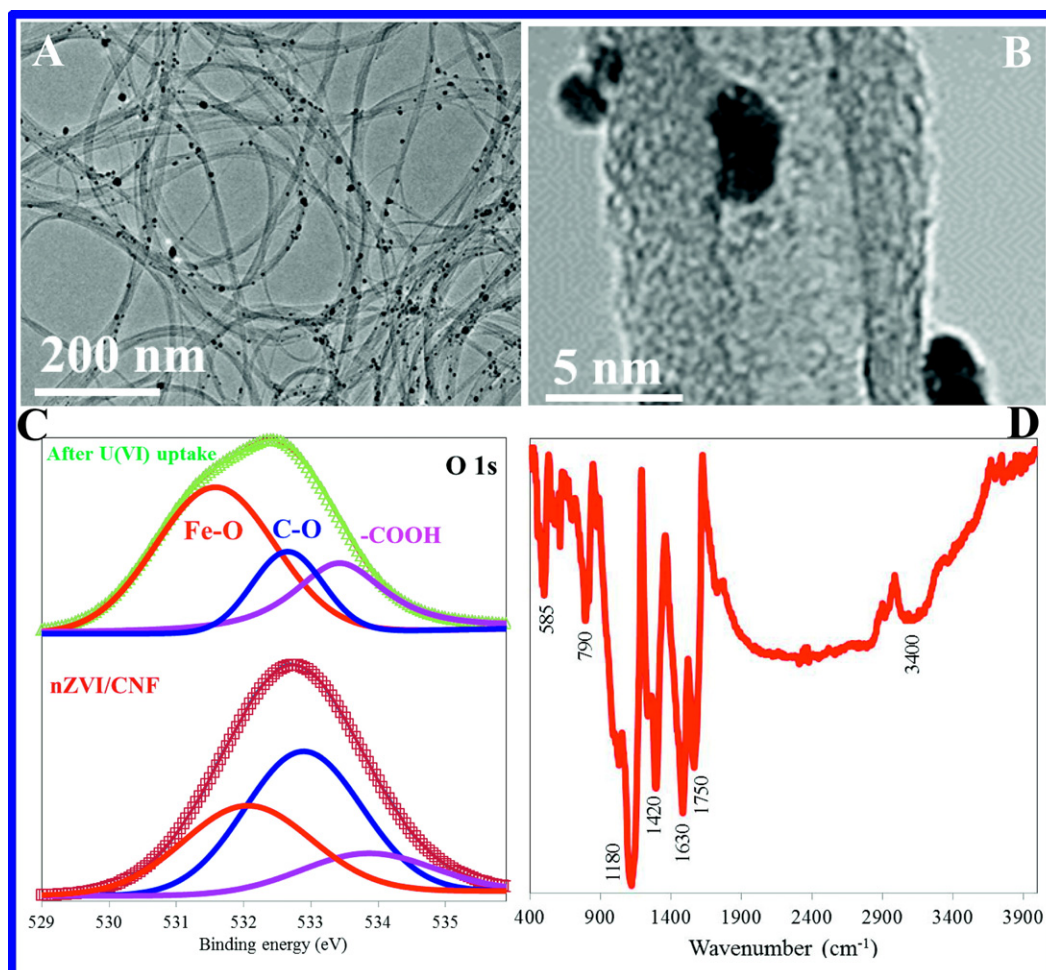


Fig. 1. Characterization of nZVI/CNF composites, A: SEM images; B: TEM image; C: O 1s XPS spectra; D: FT-IR spectra.

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