



Competitive sorption of As(V) and Cr(VI) on carbonaceous nanofibers



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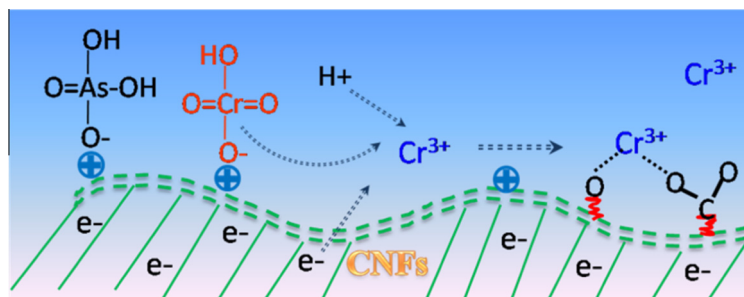
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HIGHLIGHTS

- High affinity of CNFs for Cr(VI).
- The inhibition effect of Cr(VI) on As(V) sorption on CNFs.
- The reduction of Cr(VI) to Cr(III) by CNFs.
- Dominant inner-sphere sorption of Cr(VI) on CNFs.
- Dominant outer-sphere sorption of As(V) on CNFs.

GRAPHICAL ABSTRACT



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ABSTRACT

The competitive sorption of As(V) and Cr(VI) on the novel and uniform carbonaceous nanofibers (CNFs) was investigated in single/binary-metal systems. The presence of Cr(VI) showed a greater inhibitory effect on the sorption of As(V) in binary As–Cr system, especially at higher Cr(VI) concentrations. The maximum sorption capacities of Cr(VI) and As(V) on CNFs in single-metal systems at pH 5.0 and 303 K were calculated as 2.36 and 0.67 mmol/g, respectively, by Langmuir equation. XPS results demonstrated that only As(V) species was present at the CNFs surface, whereas both Cr(VI) and Cr(III) species existed on the CNFs surfaces, suggesting the partially reduction of Cr(VI) to Cr(III) by CNFs. According to the macroscopic and spectroscopic analysis, both inner-sphere and outer-sphere surface complexation contributed to the sorption process of Cr(VI), in contrast to the electrostatic outer-sphere sorption of As(V) on CNFs. The inner-sphere Cr complexes might arise from the covalent bonding between Cr(III) and oxygen-containing functional groups of CNFs. The findings herein highlight the higher chemical affinity of CNFs for Cr(VI) relative to As(V), as a consequence of the redox reaction of the strongly toxic Cr(VI) with CNFs.

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1. Introduction

Arsenic and chromium are common contaminants in environment due to anthropogenic and biogeochemical activities [1]. Pentavalent arsenate (As(V)) exerts toxic effects on living

organisms because it can act as a phosphate analogue and inhibit the glycolytic pathway [1]. As(V) is discharged into the environments as a consequence of petroleum refining, glass melting, smelting of metal sulfide ores and natural weathering of arsenic-bearing minerals [2,3]. The elevated concentrations of chromate originate from various industries such as metallurgies, refractories and chemicals (i.e., pigments, electroplating and tanning) [4]. Hexavalent chromate (Cr(VI)) is also very toxic to biological systems because of its oxidizing, cancerogenic and corrosive properties [1]. On the contrary, trivalent chromate (Cr(III)) is less toxic and is a trace element essential for living organisms at low concentration [4]. Consequently, there is increasing public concern on removing As(V) and Cr(VI) from waters and reducing their bioavailability because of their toxicity to the environment.

Recently, novel carbonaceous nanofibers (CNFs) have gained much attention on the construction of 2D macroscopic membranes which can be applied in many fields such as optoelectronic devices [5], gas sensors [6], and filters for gas and water treatment [7]. CNFs were produced via hydrothermal carbonization (HTC) by using glucose as the precursor [8], which is green and economical and is therefore suitable for large-volume operation. In water purification, CNF membranes demonstrated efficient size-selective filtration and separation of nanoparticles from water due to the controllable diameter, narrow size distribution and high uniformity of the CNFs [9]. The high sorption capacity of CNF aerogels was also reported for the cleanup of oil spillage and chemical leakage [10]. Considering the well performance of CNFs in sewage treatment, it is significant to study their adsorption characteristics to explore a fast and highly efficient water-treatment process. The surface of the CNFs is highly functionalized with oxygen-containing groups such as –OH and C=O groups [11], thus having huge potential in removing toxic metal ions by electrostatic, complex, or hydrogen-bonding interactions. However, investigations regarding the sorption behaviors and mechanisms of toxic heavy metals on CNFs were few [11], especially in heterogeneous aquatic environments.

Herein, we studied the sorption of toxic As(V) and Cr(VI) on CNFs from aqueous solutions in this study. The objectives are: (1) to synthesize CNFs and characterize their morphology and nanostructures using scanning electron microscopy (SEM), transmission electron microscopy (TEM), X-ray photoelectron spectroscopy (XPS), and Fourier transformed infrared (FTIR) spectroscopy; (2) to investigate the individual and competitive sorption of As(V) and Cr(VI) on CNFs at different pH values and initial concentrations; and (3) to discuss the interaction mechanism of CNFs with As(V) and Cr(VI). The elucidation of the molecular-level interaction mechanisms between CNFs and heavy metal ions in multi-metal systems will provide guidance for the practical application of CNFs and CNF-based 2D macroscopic membranes in environmental pollution cleanup.

2. Materials and method

2.1. Chemicals and materials

Stock solution containing 1 mmol/L As(V) and Cr(VI) was prepared by dissolving Na₂HAsO₄·7H₂O and Na₂CrO₄ (99.9%, Sigma-Aldrich) in deionized water, respectively. The deionized water (resistivity of 18.2 MΩ cm) was obtained with a Maxima Ultrapure Water system from Elga. Glucose was supplied by Sigma-Aldrich. All other reagents in analytic grade were purchased from Sino-pharm Chemical Reagent Co., Ltd., Shanghai, China and were used directly without further purification in this study.

2.2. Preparation of CNFs

CNFs were prepared by a template-directed hydrothermal carbonization process according to previous reports [8,9,12]. Briefly, 0.6 g polyvinylpyrrolidone (PVP) and 0.0553 g Na₂TeO₃ were dissolved in 21 mL distilled water. Then 1.0 mL of hydrazine hydrate (85 w/w%) and 2.0 mL of aqueous ammonia solution were added into the solution. The final mixed solutions were transferred into a Teflon vessel (30 mL in total volume), which was closed and maintained at 180 °C for 3 h. After that, Te nanowire solution was prepared and cooled to room temperature naturally. 30 mL of acetone was added into 10 mL of as-prepared Te nanowire solution to precipitate the product before centrifuging at 6000 rpm. Then, the Te products were dispersed into 80 mL 62.5 g/L of glucose solution under vigorous magnetic stirring for 15 min. The well-defined Te@C nanocables were obtained after hydrothermal treatment of the mixed solution at 160 °C for 36 h. The pure CNFs could be obtained by chemical etching in acid H₂O₂ solution in an acid solution (HCl: H₂O₂:H₂O = 2:5:23, v/v) to remove Te cores at room temperature for 12 h. After several centrifuging–dispersing (in distilled water) cycles, the products of pure CNFs were collected. The details were provided in the previous reports [8,9,12].

2.3. Batch sorption experiments

Sorption experiments were conducted in 0.001 and 0.01 mol/L NaClO₄ solutions to investigate the sorption of As(V) and Cr(VI) on CNFs. First, NaClO₄ solutions were mixed with CNFs suspension (0.2 g/L) in 50 mL polycarbonate tubes for 24 h to keep a constant ionic strength during the experiment. Then, the stock solution of As(V) and Cr(VI) was added to the tubes with a final concentration of 0.2 mmol/L, respectively. The pH value was adjusted to be in the range 3.0–10.0 by adding a negligible volume of 1.0–0.01 mol/L NaOH and HClO₄. The sorption isotherms were investigated at pH 5.0 with As(V) and Cr(VI) concentration ranging from 0.1 to 0.6 mmol/L in 0.001 mol/L NaClO₄ solutions. Competitive sorption of As(V) and Cr(VI) on CNFs was conducted in binary-metal systems with initial concentration of As(V)/Cr(VI) = 1.0 (As–Cr system). In addition, the competitive sorption of As(V) and Cr(VI) was also performed in the presence of a 0.2 mmol/L As(V) or Cr(VI). After equilibrium, the suspension was centrifuged at 9000 rpm for 10 min and then filtered through a 0.45 μm nylon membrane filter. The concentrations of As(V) and Cr(VI) in the supernatant were measured by an inductively-coupled plasma-atomic emission spectrometer (ICP-AES, CAP6300, Thermo Scientific). To eliminate the effect of metal sorption on polycarbonate tube walls, the control experiments without adsorbents were carried out under the same experimental conditions. All experimental data were the average of triplicate determinations and the relative errors were within ±5%. The percentages and capacities of metal ion sorption on CNFs (sorption (%) and *Q* (mmol/g), respectively) were determined according to Eqs. (1) and (2), respectively.

$$\text{Sorption (\%)} = (C_0 - C_{\text{eq}})/C_0 \times 100\% \quad (1)$$

$$Q(\text{mmol/g}) = V \times (C_0 - C_{\text{eq}})/m \quad (2)$$

where *C*₀ (mmol/L) and *C*_{eq} (mmol/L) are the initial metal concentrations and the aqueous metal concentration after the sorption equilibrium, respectively, and *m* (g) and *V* (mL) represent the mass of CNFs and the volume of the suspension, respectively.

2.4. Regeneration experiment

For regeneration testing, 200 mg of CNFs exposed to 0.2 mmol/L As(V) and Cr(VI) solutions at pH 5.0 in single-metal and binary-

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