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Research article

An alternative approach for nitrate and arsenic removal from wastewater via a nitrate-dependent ferrous oxidation process

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ABSTRACT

Owing to the high efficiency of converting nitrate to nitrogen gas with ferrous iron as the electron donor, the process of nitrate-dependent ferrous oxidation (NDFeO) has been considered suitable to treat wastewater that contains nitrate but lacks organic matter. Meanwhile, arsenic immobilization often has been found during the NDFeO reaction. Thus, it was strongly expected that nitrate and arsenic could be removed simultaneously in co-contaminated wastewater through the NDFeO process. However, in the current work, arsenic was not removed during the NDFeO process when the pH was high (above 8), though the nitrate reduction rate was over 90%. Meanwhile, the biosolid particles from the NDFeO process demonstrated strong adsorption ability for arsenic when the pH was below 6. Yet, the adsorption became weak when the pH was above 7. Fourier transform infrared (FTIR) spectroscopy analysis revealed that the main activated component for arsenic adsorption was iron oxide in these particles, which was easily crippled under high pH conditions. These results implied that co-removal of nitrate and arsenic in wastewater treatment using NDFeO was difficult to carry out under high pH conditions. Thus, a two-step approach in which nitrate was removed first by NDFeO followed by arsenic adsorption with NDFeO biosolids was more feasible.

1. Introduction

Arsenic is a toxic and carcinogenic contaminant in the environment. Through mining and other human activities, a large amount of arsenic present in industrial wastewaters has been discharged into surface waters, and then transferred into the groundwater or farmland soil via irrigation, ultimately threatening human health (Marchant et al., 2017; Zhao et al., 2015). Usually, the nitrate concentration in arsenic-contaminated industrial wastewater or groundwater is relatively high (Rajmohan and Amarasinghe, 2016; Rasool et al., 2015). In fact, arsenic and nitrate are commonly reported contaminants in groundwater in many countries (Chatterjee and De, 2017). As groundwater is an important source of drinking water in some areas, pollution by arsenic and nitrate has become a serious threat to the public health in these areas (Welch and Stollenwerk, 2007). Therefore, cost-effective treatment of arsenic and nitrate in groundwater and industrial wastewater is urgently needed at present (Sarkar and Paul, 2016).

The nitrate-dependent ferrous oxidation (NDFeO) process is a combination of ferrous oxidation and nitrate reduction under anoxic conditions by specific bacteria. During the process, nitrate is mainly converted into nitrogen gas, and ferrous is oxidized into ferric iron,

which usually exists as Fe(III) (oxyhydr)oxide particles (Kappler et al., 2005; Straub et al., 1996). NDFeO bacteria have been found widely distributed in various anaerobic environments (Edwards et al., 2003; Ratering and Schnell, 2001; Weber et al., 2006a,b), of which more than 10 pure strains were isolated and reported recently (Hedrich et al., 2011). Because of the highly effective conversion of nitrate to nitrogen gas during the NDFeO reaction, NDFeO was first applied in the treatment of wastewater containing nitrate (Nielsen and Nielsen, 1998). Since then, a few technologies using this process have been developed, such as the up-flow anaerobic sludge bed and the continuous up-flow biofilter (Zhang et al., 2015; Zhou et al., 2016). Because ferrous iron costs less than adding a greater amount of organic carbon, nitrate removal based on NDFeO was considered more cost-effective than that based on heterotrophic denitrification. Moreover, this technology is suitable for nitrate removal from wastewater and polluted groundwater that lacks organic matter. In addition, arsenic immobilization via the coprecipitation or adsorption of Fe(III) (oxyhydr)oxide particles was widely observed in NDFeO reactions driven by pure strains of bacteria (Hohmann et al., 2009, 2011; Xiu et al., 2016) and in natural environments where NDFeO reactions occur (Senn and Hemond, 2002; Smith et al., 2017; Sun et al., 2016; Zhang et al., 2017). Therefore, it

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was strongly expected that nitrate and arsenic could be removed simultaneously from co-contaminated polluted water through the NDFeO process, especially for groundwater treatment (Li et al., 2015; Xiu et al., 2016).

However, during wastewater treatment based on NDFeO, the optimum pH for supporting a constant and sustained reaction might influence the adsorption of arsenic. It was reported that NDFeO is an acid-producing process (Equation (1)) (Straub et al., 1996), which results in a constant reduction in pH during wastewater treatment (Zhang et al., 2015).



Moreover, it was reported the reaction would be greatly inhibited at acidic condition (Nielsen and Nielsen, 1998). Previous research showed that only by keeping the pH near or above 8 could a high removal rate of nitrate during the wastewater treatment be guaranteed (Nielsen and Nielsen, 1998; Zhang et al., 2015; Zhou et al., 2016). Yet, the high pH conditions may impact the binding of arsenic onto Fe(III) (oxyhydr)oxide particles. Arsenic in wastewater mainly exists in anionic form, i.e., arsenate and arsenite, which are negatively charged molecules. High pH would cause the surface charge of Fe(III) (oxyhydr)oxide particles to become negative and strongly hinder the binding of arsenic (Ociński et al., 2016). Hence, it is doubtful that arsenic co-removal could be carried out if high pH remained during the NDFeO process. Obviously, an alternative approach for the treatment of nitrate and arsenic co-polluted wastewater needs to be developed when the conflict among optimal conditions occurred.

Thus, for wastewaters from which arsenic and nitrate could not be co-removed using the NDFeO process in a single reactor, a two-step approach based on NDFeO was hypothesized and tested in this work. The flow diagram of the two-step approach is represented in Fig. S1. “Two-step” means two reactors are required: one for nitrate reduction using the NDFeO process and one for arsenic removal via adsorption employing the biosolids formed in the NDFeO process. As just mentioned, the Fe(III) (oxyhydr)oxide particles produced via the NDFeO process are capable of binding arsenic. These particles often aggregate together on the surface of bacterial cells, becoming the biosolids (sludge) in the biological treatment of wastewater (Li et al., 2017; Zhang et al., 2015), and thus would be potential adsorbents. Hence, it was hypothesized that they could be collected for use in an additional adsorption step targeting the arsenic if arsenic co-removal did not occur during wastewater treatment using NDFeO to reduce nitrate. Thus, the removal of nitrate and arsenic based on the NDFeO process could be achieved. However, the properties of these biosolids as arsenic adsorbents has not been a concern of the research to date.

Thus, the aims of this study were as follows: 1) to verify whether the NDFeO process could accomplish arsenic co-removal under high pH conditions (near 8), 2) to determine the properties of arsenic adsorption by biosolids generated during the NDFeO process, and 3) to consider the possibility of arsenic removal via adsorption by these biosolids.

2. Material and methods

2.1. Bacterial enrichment

Mixed NDFeO bacteria from the sediments of the Chebei River in the city of Guangzhou were obtained via enrichment culture (Xie et al., 2015). Our previous study had shown that these bacteria could regulate the NDFeO reaction quickly when the concentration of ferrous was about 2–3 mmol/L, which is the concentration of ferrous in the sediment interstitial water (Fig. S2).

The enrichment culture was routinely cultivated in anoxic heterotrophic denitrification medium (Xie et al., 2015) that contained 267.5 mg/L of NH_4Cl , 100 mg/L of KH_2PO_4 , 100 mg/L of K_2HPO_4 , 310 mg/L of NaNO_3 , 160 mg/L of CH_3COONa , and 1 mL/L trace element solution at pH 6.8–7.2. The trace element solution contained

6.37 g/L of Na_2EDTA , 2.2 g/L of ZnSO_4 , 5.5 g/L of CaCl_2 , 5.06 g/L of $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$, 5.0 g/L of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$, 1.1 g/L of $\text{Na}_2\text{MoO}_4 \cdot 4\text{H}_2\text{O}$, 1.57 g/L of $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$, and 1.61 g/L of $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$. All growth experiments were performed at 30 °C in the dark and were not shaken.

2.2. Mixed-bacteria-driven NDFeO process with different arsenic concentrations

The NDFeO bacterial culture medium was prepared by modifying the heterotrophic denitrification culture medium mentioned in Section 2.1: FeCl_2 was added and the ferrous concentration was controlled to 2 mmol/L (112 mg/L), whereas the nitrate-N concentration was adjusted to about 4 mmol/L (56 mg/L). As a co-electron donor for NDFeO, acetate with initial concentration of 2 mmol/L (160 mg/L) was added. A solution containing As was prepared by adding $\text{Na}_3\text{AsO}_4 \cdot \text{H}_2\text{O}$ to the NDFeO medium for two arsenic concentrations of 0.1 and 0.5 mg/L. Moreover, NaHCO_3 was used as a buffering agent for pH adjustment, and based on our previous experiment, the NaHCO_3 concentration was controlled at 168 mg/L to maintain the pH near 8.

A series of 2.5 L four-hole reaction flasks were used as reactors. NDFeO experiments were carried out by inoculating cells of the enrichment culture into the two media containing arsenic. In each experiment, 500 mL of enrichment culture was centrifuged for cell retrieval (5000 r/min, 10 min). The cells were resuspended in sterilized medium. Into a reactor containing 2 L sterilized medium, 100 mL of the cell resuspension was added. The reaction flask was filled with N_2 for 30 min to obtain the anoxic conditions and then cultivated in a 30 °C incubator. Each arsenic concentration system had a corresponding control system, which lacked bacteria. Each treatment was examined in triplicate. Sampling and the measurement of pH were performed at regular intervals in each reaction system, with 15 mL samples removed each time. 5 mL of the samples was placed in vials containing 0.5 mol/L hydrochloric acid for overnight to extract ferrous ion. The other 10 mL of liquid was separated via filtration using 0.45 μm membrane filters. The concentrations of $\text{NO}_3\text{-N}$, acetate, and arsenic were determined after filtration.

2.3. Characteristics of arsenic adsorption by biosolids generated in the NDFeO reaction

2.3.1. Preparation of the biosolids from NDFeO

For the biosolid formation, a 10 L flask sealed with a rubber stopper with gas-guide tubes was used as the reactor, into which 9.5 L of NDFeO medium and 0.5 L of enrichment culture were added. Specific steps and details of the biosolid formation process using NDFeO were described in our previous work (Li et al., 2017). Finally, solid particles were obtained for the arsenic adsorption experiments that followed.

2.3.2. Arsenic adsorption experiments

Adsorption experiments were carried out under a batch regime, using a thermostatic shaker at 180 rpm and 30 °C. Each one was performed in triplicate. A series of equilibrium experiments was first conducted in 100 mL conical flasks with 0.2 g adsorbent (the biosolids) and 80 mL of $\text{Na}_3\text{AsO}_4 \cdot \text{H}_2\text{O}$ solution. Because the adsorption is more applicable for treating wastewater with low arsenic concentration, the initial level of arsenic was designated in the range of 0.05–0.5 mg/L. Five solutions with different pH values (4, 5, 6, 7, and 8) were prepared for each initial concentration. For each adsorption experiment, the residual arsenic in solution was measured after 48 h. The adsorption kinetic experiment was carried out in a 250 mL conical flask with 1 g of adsorbent and 200 mL of solution and was performed under the pH condition with the highest equilibrium removal rate. At given intervals, the concentration of arsenic in solution was determined.

Fourier transform infrared (FTIR) spectroscopy (Bruker Vertex 70, German) and X-ray photoelectron spectroscopy (XPS) (Shimadzu Co., Japan) analysis of the biosolids was performed before and after the

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