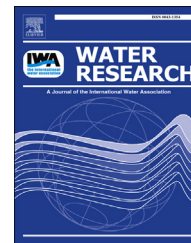


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# Modification of ultrafiltration membrane with nanoscale zerovalent iron layers for humic acid fouling reduction

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## ABSTRACT

Nanoscale zerovalent iron (NZVI) was layered onto ultrafiltration (UF) membrane surface and tested for antifouling properties using humic acid (HA). Scanning electron microscopy showed that a relatively homogeneous layer was formed across the membrane surface by NZVI particles. Strong adhesion was observed between NZVI and UF membrane used. HA was significantly removed and membrane flux was increased in the presence of NZVI layer. Increased loadings of NZVI onto the membrane surface increased resistance to fouling while slightly reducing the clean water permeability of the membrane. However, the pore size of the layer formed by pristine NZVI was large, resulting in more chances of HA molecules getting to the membrane surface even blocking the membrane pores at the beginning. Membrane loaded with NZVI layer performed much better under acidic conditions. During NZVI synthesis, specific surface area of NZVI particle increased with increasing the ratio of ethanol ( $V_{\text{ethanol}}/V_{\text{solution}}$ ), which also gradually decreased the average pore size of NZVI layer. As a result, the corresponding membrane flux steadily increased. Additionally, the results for permeate samples under different conditions showed that large molecular weight (MW, >30 kDa) and medium MW HA molecules (3–30 kDa) were removed much faster than those of small MW HA molecules (<3 kDa).

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## 1. Introduction

Humic substances (HS) can form during the decomposition of plant/animal biomass in natural systems and are some of the most abundant materials on earth. HS comprise a skeleton of alkyl and aromatic units with different functional groups, such as carboxylic acid, phenolic hydroxyl, ketone, and

quinone groups (Giasuddin et al., 2007). The typical molecular weight (MW) of HS ranges from a few thousand to a few hundred thousand daltons with a dependence on their source (Yuan and Zydney, 1999). HS are commonly existed in natural waters in the range of a few mg/L to a few hundred mg/L C (Wall and Choppin, 2003). However, the presence of HS could cause various environmental and health problems including

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the followings: (1) HS could cause undesirable color and taste, which are also served as food for bacterial growth in water (Bai and Zhang, 2001); (2) HS could bind with heavy metals and biocides, yielding high concentration of these substances and enhancing their transport in water (Schmitt et al., 2003); (3) HS could react with chlorine during water treatment to form disinfection by-products, such as trihalomethane (Tian et al., 2013); (4) HS have been shown to compete with low MW synthetic organic chemicals and inorganic pollutants, reducing their adsorption rates and equilibrium capacities (Tratnyek et al., 2001; Klausen et al., 2003); (5) HS could act as a major foulant causing serious microfiltration (MF)/ultrafiltration (UF) membrane fouling during filtration (Yuan and Zydney, 1999, 2000). As such, the removal of HS has been widely investigated to minimize their impact on environmental and human health.

Membrane processes have been increasingly used in water treatment, mainly owing to the superior and stable effluent quality even under conditions with variable feed-water characteristics (Guo et al., 2010). However, membrane fouling is inevitable and is still a major impediment to broader application of membrane technology (Ajmani et al., 2012). To better control membrane fouling, at least three processes have been investigated to combine adsorption and UF membrane filtration: (1) pre-adsorption, in which the feed solution is contacted with and then the adsorbent is separated before water is applied to the membrane; (2) direct filtration, in which the adsorbent and feed solution are mixed together and then applied directly to the membrane; and (3) pre-deposition, in which an adsorbent layer is deposited onto the membrane surface before the application of feed solution (Cai et al., 2013). The first two processes have been applied successfully in many full-scale treatments, while the last has only been tested at laboratory-scale (Thiruvengkatachari et al., 2006; Ajmani et al., 2012; Ma et al., 2013). Previous studies show that heated iron oxide particles (HIOPs) performed better on reducing membrane fouling than those of powdered activated carbon (PAC) and SiO<sub>2</sub> particles when they are layered onto the membrane surface (Zhang et al., 2003a,b). Pre-deposited HIOPs and heated aluminum oxide particles (HAOPs) are also more effective to remove NOM than that of PAC, which preferentially adsorbs many non-fouling NOM molecules (Kim et al., 2008). According to the results of Ajmani et al. (2012), carbon nanotubes (CNTs) layering low pressure membranes are highly resistant to membrane fouling, especially when the diameter of CNTs ranges from 50 to 80 nm. In our previous study, iron/aluminum hydrolyzed precipitate layer has better antifouling properties, especially for aluminum hydrolyzed precipitate layer (Ma et al., 2013). Overall, these laboratory-scale tests have generally shown that pre-deposition could yield excellent results to some extent, which is even better than the other two with respect to both organic removal and membrane fouling control (Kim et al., 2010; Cai and Benjamin, 2011).

In comparing with zerovalent iron (ZVI)/iron oxide, there is a larger adsorption capacity of organic/inorganic matter to nanoscale zerovalent iron (NZVI) to some extent (Zhang et al., 2003a,b; Giasuddin et al., 2007; Du et al., 2013; Fu et al., 2014; Wu et al., 2014). NZVI is firstly synthesized by adding NaBH<sub>4</sub> solution dropwise to a FeCl<sub>3</sub>·6H<sub>2</sub>O solution with magnetic

stirring (Wang and Zhang, 1997). The larger specific surface area and higher surface activity of NZVI has interested many researchers (Lowry and Johnson, 2004). NZVI has most frequently been applied in groundwater remediation by injecting its water-particle slurry into the contaminated area (Zhang and Elliott, 2006). A wide array of environmental common contaminants could be transformed/adsorbed by NZVI, such as de-chlorination of chlorinated organic compounds (Liu et al., 2005), reduction of nitrate (Jiang et al., 2011), degradation of trichloroethylene (Zhang et al., 2011), adsorption of Cr (VI) and Pb (II) (Ponder et al., 2000). Up to now, NZVI is used in several field tests and has shown promising prospective for in-situ remediation (Li et al., 2006b; Zhang and Elliott, 2006). Over the last few years, various methods have been developed for the production of NZVI particles (Li et al., 2006a, 2006b) and the modification of their surface properties (Liu et al., 2005; Li et al., 2013). Recently, NZVI has been introduced into water treatment processes due to its high reactivity and large specific surface area. It shows that kaolin supported NZVI is efficient at Pb (II) removal from electroplating wastewater, and Cr (VI) could largely be removed by bentonite-supported NZVI from wastewater (Zhang et al., 2010; Shi et al., 2011). Despite the great potential of NZVI particles, little focus has been placed on the investigation of membrane technology with NZVI particles.

Herein, NZVI particles were applied before UF membrane filtration in this paper. The primary objective of this research is to develop a NZVI layered membrane that can exhibit improved resistance to fouling from HS in comparison with the pristine membrane. As part of this study, some factors which were responsible for determining the antifouling properties of NZVI layers were investigated, such as doses, solution pH, specific surface area of NZVI particles, pore size and thickness of the layer, etc. Finally, the adhesion ability of NZVI to UF membrane was investigated in detail and the MW distributions of the permeate samples under different conditions were analyzed.

## 2. Materials and methods

### 2.1. Materials

The chemical reagents used in this study, such as NaCl, FeCl<sub>3</sub>·6H<sub>2</sub>O, NaBH<sub>4</sub>, HCl, NaOH, and ethanol were analytical grade obtained from Sinopharm Chemical Regent Co., Ltd (China). Deionized (DI, Millipore Milli-Q) water was used in the experiments. Humic acid sodium salt (HA, Sigma–Aldrich, U.S.A), a representative of HS, was dissolved by DI water followed by filtering the solution through a 0.45 μm membrane (Cellulose Acetate). All chemical stock solutions were stored in the dark at 4 °C.

Nanoscale zerovalent iron was synthesized following the method reported by Wang and Zhang (1997), in which NaBH<sub>4</sub> solution was added dropwise to a FeCl<sub>3</sub>·6H<sub>2</sub>O solution with magnetic stirring. In order to get a higher specific surface area of NZVI particles, different ratios of ethanol ( $V_{\text{ethanol}}/V_{\text{solution}}$ ) were used instead of 100% water (Liu et al., 2005). Finally, DI water was used to remove the dissolved salts by repeated rinsing of the solids three times.

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