



Removal of dissolved organic matter and control of membrane fouling by a hybrid ferrihydrite-ultrafiltration membrane system

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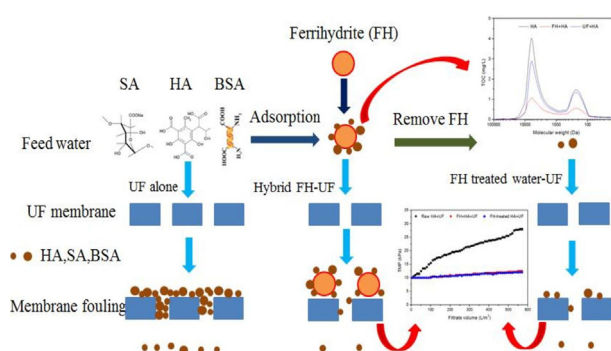
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HIGHLIGHTS

- FH exhibited a larger adsorption capacity than PAC for each DOM fraction.
- The hybrid FH-UF process effectively removed DOM and mitigated membrane fouling.
- The hybrid PAC-UF process with PAC particles didn't alleviate HA and SA fouling.
- The two hybrid adsorption membrane processes significantly reduced BSA fouling.
- Both calcium ions and the magnetism of FH contributed to a loose FH layer.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 30 November 2017

Received in revised form 13 February 2018

Accepted 5 March 2018

Available online xxxx

Editor: Zhen (Jason) He

Keywords:

Ultrafiltration (UF)

Membrane fouling

Dissolved organic matter (DOM)

Ferrihydrite (FH)

Adsorption

ABSTRACT

This study investigated the pretreatment effect of ferrihydrite (FH) on the removal of dissolved organic matter (DOM) and membrane fouling control in a hybrid adsorption-ultrafiltration (UF) membrane system. Powdered activated carbon (PAC) was chosen for comparative study, and three surrogates, namely, humic acid (HA), sodium alginate (SA) and bovine serum albumin, were applied in feed water to simulate humic substances, polysaccharides and proteins, respectively. In the hybrid adsorption-membrane system, the impacts of adsorbent cake layers on DOM removal and membrane fouling were systematically investigated. The results demonstrated that FH effectively removed both high-molecular-weight (MW) and low-MW HA and SA molecules via ligand exchange and mitigated membrane fouling substantially, regardless of whether FH particles were removed from the feed water before UF. In addition, the large flocs induced by FH and calcium contributed to loose FH layers. In contrast, PAC had almost no capacity to adsorb high-MW DOM molecules due to the microporous structure. And FH was more efficient in removal of DOM and control of membrane fouling than PAC. The different DOM adsorption capacities of FH and PAC lead to different effects on membrane fouling caused by deposition of DOM molecules and cake layers. These results offer guidance for application of hybrid adsorption-membrane filtration systems.

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1. Introduction

The low-pressure ultrafiltration (UF) membrane is a promising approach used in water treatment processes because of its low energy

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consumption, lack of chemical addition and high removal efficiency for particulate organic matter, turbidity and microorganisms compared with the conventional treatment technologies (Acero et al., 2012; Ang et al., 2006). However, a UF membrane with a relatively large pore size in the range of 0.01–0.1 μm is not effective in removing dissolved organic matter (DOM) (Hill et al., 2009; Kwon et al., 2005). DOM, which includes humic substances (HS), proteins and polysaccharides, is a main concern in drinking water production because it is considered a precursor of disinfection by-products (DBPs) (Krasner et al., 2006). In addition, membrane fouling caused by the accumulation of organic matter on the membrane surface induces higher production costs and limits the more widespread application of UF membranes (Lohwacharin et al., 2010; Ochando Pulido, 2016; Shao et al., 2016). Therefore, proper pretreatment for minimization of fouling and improved DOM removal is essential for successful UF operation.

Adsorption is an efficient technology for DOM removal and is widely used as a pretreatment for UF to enhance the quality of treated water and reduce membrane fouling (Huang et al., 2009; Margot et al., 2013; Sheng et al., 2016). Powdered activated carbon (PAC) is considered as a good adsorbent because of its well-developed internal pore structure, large surface area and many functional groups (Heidari et al., 2014) resulting in a large adsorption capacity. Therefore, it was investigated as a pretreatment in the hybrid adsorption/UF process (Stoquart et al., 2012). Although PAC always removed some DOM and enhanced the quality of treated water, its impact on membrane fouling is still under debate (Stoquart et al., 2012). It was reported that PAC adsorbed a large amount of DOM, which increased the membrane permeability and reduced the membrane fouling (Osawa et al., 2017). However, in other studies, PAC adsorbents were reported to exert negative effects on fouling control due to accumulated dense cake formation (Chu et al., 2017). Although PAC has the ability to reduce the fouling caused by DOM, it also contributed to the formation of a dense cake layer, leading to an overall increase in fouling (Zhang et al., 2003).

Ferrihydrite (FH) is an amorphous iron oxide particle (IOP) that has high reactivity, a large specific surface area and a high adsorption capacity for arsenate, phosphate and DOM (Antelo et al., 2010). Both crystallized (e.g., hematite and goethite) and amorphous IOP can adsorb DOM via ligand exchange between the acidic carboxylic and the phenolic groups of DOM and the hydroxyl groups of IOP to form inner-sphere complexes or via physisorption to form outer-sphere complexes (Gu et al., 1994; Ha et al., 2008). Lin et al. (2001) proposed that HS significantly contributed to flux decline as manifested in several studies showing that humic acid (HA) is the main material in organic fouling (Chu et al., 2017; Shao et al., 2016). FH might remove HA efficiently via ligand exchange and reduce membrane fouling (Yang et al., 2014). Kang and Choo (2010) used FH as a pretreatment for the UF membrane, and their results showed that large-MW DOM was effectively removed by FH adsorption, which can be retained on the membrane surface and deteriorate membrane fouling on the UF membrane. In addition, other researchers also reported that the strong bonding of DOM molecules to FH resulted in the loose FH cake layer, which could reduce membrane fouling (Weerasekara and Choo, 2015; Zhang et al., 2003). Therefore, FH adsorbents as pretreatment have a potential to control membrane fouling in the hybrid membrane system. Even so, the performances of FH in removal of DOM with different fractions and the influence on membrane fouling caused by different compositions are still unclear. The relevant underlying mechanisms also remained to be studied. In addition, the structure of FH cake layer and its formation mechanism has not been clearly explained. In the different compositions of DOM, a polysaccharide-like compound and protein-like substances has been reported to exert large influence on membrane fouling (Li et al., 2014; Zhou et al., 2014). Therefore, three model foulants of HA, bovine serum albumin (BSA) and sodium alginate (SA) were used in feed water to represent different DOM fractions (Li et al., 2014; Shao et al., 2017) for evaluation of whether FH could effectively remove DOM fractions and reduce membrane fouling in the hybrid membrane system.

The aim of this study was to obtain a comprehensive understanding of the effects of FH pretreatments on DOM removal and UF membrane fouling control. PAC was chosen for comparative study. The adsorption capacities of FH and PAC towards three foulants, namely, HA, BSA and SA, were evaluated, and cross-flow UF experiments were performed with and without adsorption pretreatment. Moreover, the effect of adsorbent particles that formed a cake layer on the removal efficiencies of DOM and membrane fouling were also examined. Membrane fouling is reflected by the changes in the TMP, and the fouling resistance was calculated using the resistance-in-series model.

2. Materials and methods

2.1. DOM solutions

HA, SA and BSA purchased from Sigma-Aldrich (USA) were used as representatives of HS, proteins and polysaccharides, respectively. To prepare the HA stock solution, HA was dissolved in Milli-Q water, and the solution pH was adjusted to 7.5 by addition of NaOH under mixing with a magnetic stirrer. The solution was subsequently filtered through a 0.45- μm filter. The stock HA solution was stored at 4 °C in the dark until use. The SA and BSA stock solutions were prepared in the same manner and were diluted with Milli-Q water before adsorption and membrane experiments.

Considering the dissolved organic carbon (DOC) in the real water, the concentrations of HA, BSA and SA used in UF experiments were all set to 5.0 mg-C/L. To simulate the solution chemistry of natural water, 6 mmol/L NaCl, 1 mmol/L NaHCO₃ and 1 mmol/L CaCl₂ were added, and the pH was adjusted to 7.5 with 0.1 mol/L HCl and NaOH (Li et al., 2014).

2.2. Adsorbents and adsorption experiments

2.2.1. Preparation and characterization of FH and PAC

Two adsorbents of FH and PAC were used in this study. The 2-line FH was synthesized in the laboratory according to the method of Leone (Leone et al., 2001). The concentration of FH was measured by inductively coupled plasma atomic emission spectroscopy (ICP-AES, ULTIMA, JY, France), if necessary, after dilution. The PAC (Xilong, China) chosen for comparative study was cleaned prior to use according to the reference (Lohwacharin et al., 2010).

The BET surface area and the pore size distributions of FH and PAC were characterized using a specific surface and porosity analyzer, as shown in Table 1. The average pore size of the FH particles was 4.5 nm, which was larger than the 1.9 nm of PAC adsorbent.

2.2.2. Adsorption kinetics and isotherm tests

Experiments on the adsorption kinetics and equilibrium on FH and PAC were performed with the bottle-point technique. All batch adsorption experiments were conducted with 100 mL of water samples at 25 °C during shaking at 100 rpm. For the kinetic test, 50 mg/L adsorbents were dosed into solutions and reacted from 5 to 90 min while various amounts of adsorbents were added to DOM solution for 30 min for the isotherm experiment. The water samples were filtered immediately through a 0.45- μm filter to remove adsorbent particles for DOC analysis.

Table 1
Characteristics of the adsorbents used.

Parameters	FH	PAC
Average particle size (μm)	6.88	8.45
Surface area (m^2/g)	289	836
Average pore size (nm)	4.5	1.9
Pore volume (cm^3/g)	0.189	1.36

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