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Comparison of the effects of aluminum and iron(III) salts on ultrafiltration membrane biofouling in drinking water treatment

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ABSTRACT

Coagulation plays an important role in alleviating membrane fouling, and a noticeable problem is the development of microorganisms after long-time operation, which gradually secrete extracellular polymeric substances (EPS). To date, few studies have paid attention to the behavior of microorganisms in drinking water treatment with ultrafiltration (UF) membranes. Herein, the membrane biofouling was investigated with different aluminum and iron salts. We found that $Al_2(SO_4)_3{\cdot}18H_2O$ performed better in reducing membrane fouling due to the slower growth rate of microorganisms. In comparison to $Al_2(SO_4)_3{\cdot}18H_2O$, more EPS were induced with $Fe_2(SO_4)_3{\cdot}xH_2O$, both in the membrane tank and the sludge on the cake layer. We also found that bacteria were the major microorganisms, of which the concentration was much higher than those of fungi and archaea. Further analyses showed that *Proteobacteria* was dominant in bacterial communities, which caused severe membrane fouling by forming a biofilm, especially for $Fe_2(SO_4)_3{\cdot}xH_2O$. Additionally, the abundances of *Bacteroidetes* and *Verrucomicrobia* were relatively higher in the presence of $Al_2(SO_4)_3{\cdot}18H_2O$, resulting in less severe biofouling by effectively degrading the protein and polysaccharide in EPS. As a result, in terms of microorganism behaviors, Al-based salts should be given preference as coagulants during actual operations.

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Introduction

Ultrafiltration (UF) membranes have been widely used in drinking water treatment due to the improved quality of effluent produced, even with variable feed-water properties (Jermann et al., 2007). However, membrane fouling is inevitable after long-time operation, which has constrained its further utilization (De Souza and Basu, 2013). Previous studies have shown that membrane fouling leads to the reduction of membrane flux and an increase in energy consumption, resulting in increased cost during water treatment (Leiknes, 2009).

Three main fouling mechanisms are known to occur as a function of time: pore constriction, pore blocking and cake layer formation (Wang and Tarabara, 2008). To effectively alleviate membrane fouling, various kinds of technologies have been investigated, such as coagulation, adsorption, preparing new membrane materials (Dong et al., 2007; Hua et al., 2008; Gong et al., 2015), *etc.* However, owing to its lower cost, easier operation procedure, and higher removal

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efficiency for pollutants, coagulation is still the most promising method in alleviating membrane fouling currently, and it has already been widely applied in water plants (Gao et al., 2011).

For coagulation, Al-based salts and Fe-based salts are the most commonly used coagulants. To reduce membrane fouling, a number of studies have paid much attention to improving the removal efficiency, and it has been demonstrated that the hydrolyzed flocs play an important role (Jiang and Graham, 1996; Jeong et al., 2014; Wang et al., 2017). Further research has revealed that aluminum speciation and iron speciation are also critical to the removal efficiency, especially aluminum speciation (Zhao et al., 2009; Ma et al., 2015). It has been shown that monomeric aluminum species are easily bound to polysaccharide and cellulosic molecules (Masion et al., 2000; Dong et al., 2014). The preferred species Al_{13} are easily bound to carboxylic groups under acidic conditions, while they are easily bound to phenolic moieties under alkaline conditions (Kazpard et al., 2006).

In recent years, the removal mechanism after coagulation has gradually become much clearer. However, a noticeable problem is the development of microorganisms after long-time operation, which can produce extracellular polymeric substances (EPS). These EPS, mainly composed of protein and polysaccharide, not only cause severe membrane fouling by forming a denser biofilm, but also can deteriorate the effluent quality (Komlenic, 2010). Most studies have focused on inhibiting the development of microorganisms through disinfection (Komlenic, 2010; Gao et al., 2011; Hook et al., 2012), while the composition and function of the microbial community in the cake layer are largely unknown.

Herein, to fully understand the membrane biofouling after coagulation, the membrane performance was investigated with $Al_2(SO_4)_3{\cdot}18H_2O$ and $Fe_2(SO_4)_3{\cdot}xH_2O$ because of the strong corrosiveness of $FeCl_3{\cdot}6H_2O$ during actual operations (Esih et al., 2005). The purpose is to provide a better understanding of the influence of coagulants on microorganisms' behaviors, including the growth of microorganisms, the proportion of protein and polysaccharide in EPS, and the membrane fouling contributed by bacteria, fungi, archaea, *etc.*

1. Materials and methods

1.1. Materials

The chemical reagents used were analytical grade except where specified. $Al_2(SO_4)_3{\cdot}18H_2O$ and $Fe_2(SO_4)_3{\cdot}xH_2O$ were purchased from Sinopharm Chemical Regent, Co., Ltd. (China). To simulate micro-polluted surface water, domestic sewage was mixed with tap water, at a volume ratio of 1:50 (Yu et al., 2014). The characteristics of the feed water are shown in Table 1.

Table 1 – Characteristics of feed water.

Parameters	Feed water
pH	7.52 ± 0.11
Water temperature (°C)	21.8 ± 1.7
Turbidity (NTU)	2.97 ± 0.08
Average particle size (nm)	21.7 ± 4.8
NO_2^- (mg/L)	0.64 ± 0.11
NO_3^- (mg/L)	3.62 ± 0.45
NH_4^+ (mg/L)	1.32 ± 0.24
UV_{254} (cm^{-1})	0.14 ± 0.01
Total organic carbon (TOC, mg/L)	4.09 ± 0.26

1.2. Experimental setup

Fig. 1 shows the schematic diagram of the experimental setup. For the coagulation section, the concentration of $Al_2(SO_4)_3{\cdot}18H_2O$ or $Fe_2(SO_4)_3{\cdot}xH_2O$ was 0.05 mmol/L. A rapid mixing speed was maintained at 300 r/min for 1 min, and then decreased to 100 r/min for 14 min. For the filtration section, a polyvinylidene fluoride (PVDF) hollow fiber membrane (Motianmo, China) was used, and the average pore size was 30 nm (provided by the manufacturer).

The total surface area of the submerged membrane in the membrane tank was 0.025 m^2. The constant permeate flux was kept at 20 L/(m^2·hr), with a cycle of filtration for 30 min followed by 1 min backwashing (40 L/(m^2·hr),) with aeration (100 L/hr). The hydraulic retention time (HRT) of the membrane tank was 0.5 hr. During the operation, the transmembrane pressure (TMP) was monitored each day to reflect the development of membrane fouling. No additional disinfection method was used during filtration and the sludge was discharged every three days.

1.3. Characteristics of flocs

To investigate the membrane performance in detail, floc characteristics were tested with a jar test. The beaker (1.0 L) was linked with a Mastersizer 2000 laser diffraction instrument (Malvern, UK) by a silicone tube (internal diameter: 5 mm). The water was driven by a suction peristaltic pump at a flow rate of 2 L/hr (Yu et al., 2015). For the test, the rapid mixing speed was also maintained at 300 rpm for 1 min, and then decreased to 100 r/min for 14 min. D_{50} was used to represent the average diameter of flocs. The fractal dimension (D_f) was calculated with the small angle light scattering method when flocs reached their steady-state size (Wu et al., 2002).

1.4. Measurement of EPS in cake layer and membrane tank

The foulants on the membrane surface were washed by phosphate buffer saline solution (0.01 M, pH 7.4) after filtration. Then, the solution was heated at 80°C for 30 min, followed by centrifuging at 20,000 r/min for another 5 min, and then the supernatant was collected for EPS analysis (Zhang et al., 1999). The concentration of protein was determined using a BCA kit (Tiangen, China), and the concentration of polysaccharide was measured by the phenol-sulfuric acid method (Saha and Brewer, 1994). Similar methods were also employed for measuring the concentration of protein and polysaccharide in the water of membrane tank.

1.5. Microscopic observation of fouling layer

Five centimeters of membrane was cut from the membrane modules at the end. These membrane fibers were placed in 0.1 mol/L phosphate buffer with 3.0% glutaraldehyde at pH 7.2

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