JOURNAL OF ENVIRONMENTAL SCIENCES XX (2018) XXX–XXX

Available online at www.sciencedirect.com

ScienceDirect

www.elsevier.com/locate/jes

JES JOURNAL OF ENVIRONMENTAL SCIENCES

www.jesc.ac.cn

The effects of hydrogen peroxide pre-oxidation on ultrafiltration membrane biofouling alleviation in drinking water treatment

Xing Wang[1,2], *Baiwen Ma*[1], *Yaohui Bai*[1,*], *Huachun Lan*[1], *Huijuan Liu*[1], *Jiuhui Qu*[1]

1. *Key Laboratory of Drinking Water Science and Technology, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China*
2. *University of Chinese Academy of Sciences, Beijing 100049, China*

ARTICLE INFO

Article history:
Received 22 November 2017
Revised 18 January 2018
Accepted 19 January 2018
Available online xxxx

Keywords:
Ultrafiltration membrane
Hydrogen peroxide
Fouling alleviation
Extracellular polymeric substances
Microbial community

ABSTRACT

Pre-oxidation is widely used to reduce ultrafiltration membrane fouling. However, the variation in the composition of microbial communities and extracellular polymeric substances (EPSs) accompanying pre-oxidation in drinking water treatment has received little attention. In this study, hydrogen peroxide (H_2O_2) was used in a coagulation-ultrafiltration process with $Al_2(SO_4)_3{\cdot}18H_2O$. A long-term reactor experiment (60 d) showed that pre-oxidation alleviated membrane fouling, mainly due to its inhibition of microbial growth, as observed by flow cytometry measurements of the membrane tank water. Further analysis of the formed cake layer demonstrated that the corresponding levels of EPS released from the microbes were lower with than without H_2O_2 treatment. In comparison to polysaccharides, proteins dominated the EPS. 2D-electrophoresis showed little difference ($p > 0.05$, Student's *t*-test) in the composition of proteins in the cake layer between the treatments with and without H_2O_2. The molecular weights of proteins ranged from approximately 30–50 kDa and the majority of isoelectric points ranged from 6 to 8. High-throughput sequencing showed that the predominant bacteria were *Proteobacteria*, *Bacteroidetes*, and *Verrucomicrobia* in both cake layers. However, the relative abundance of *Planctomycetes* was higher in the cake layer with H_2O_2 pre-oxidation, which was likely probably due to the strong oxidative resistance of its cell wall. Overall, our findings clarify the fundamental molecular mechanism in H_2O_2 pre-oxidation for ultrafiltration membrane bio-fouling alleviation in drinking water treatment.

Introduction

Ultrafiltration membranes are widely used in drinking water treatment due to their excellent characteristics, such as easy modularization, land use reduction, and effluent water improvement (Laîné et al., 2000). However, membrane fouling is inevitable after long-term operation, which has, to some extent, constrained their utilization. Previous studies have demonstrated that membrane fouling can lead to a reduction in membrane flux and an increase in energy consumption, thereby increasing the cost of water treatment (Sheng et al., 2010).

* *Corresponding author. E-mail:* yhbai@rcees.ac.cn (Yaohui Bai).

https://doi.org/10.1016/j.jes.2018.01.020
1001-0742/

Membrane fouling includes inorganic, organic, colloidal, and biofouling (Pearce, 2007; Zhang et al., 2009; Luo et al., 2015; Abdelrasoul et al., 2017). In comparison to other fouling mechanisms, biofouling gradually becomes the predominant fouling mechanism, especially after long-term operation. It is always induced by the formation of a biofilm due to the adhesion of microorganisms and their metabolites on the membrane surface (Zhou et al., 2007; Gong et al., 2015). In addition to flux decline and energy consumption increase, biofouling also has adverse effects regarding water treatment. The formation of biofilms, which release extracellular polymeric substances (EPSs), can lead to severe fouling and deterioration in the quality of treated water (Wimpenny et al., 2000; Komlenic, 2010). In addition, the acidic by-products produced by microorganisms can degrade membranes (Murphy et al., 2001). Therefore, biofouling control/alleviation is a critical issue in membrane filtration processes. To date, anti-biofouling compounds, such as silver salts, nitrofurazone, and ammonium surfactants, have been used to mitigate membrane biofouling for water treatment (Zhang et al., 2011; Hook et al., 2012; Kaegi et al., 2013). However, these compounds, which can increase the effluent water quality risk due to their small molecular weight (so that they pass through the membrane into the effluent), are not suitable for drinking water treatment.

As a traditional method for the treatment of drinking water, pre-oxidation can alleviate membrane fouling by inhibiting the growth of microorganisms and altering the composition of EPS (Gao et al., 2011). In comparison with chemically enhanced backwash, pre-oxidation is easy to use, provides continuous effects on inhibiting microbial growth and causes less damage to membranes (Chen et al., 2003; Cai and Liu, 2016). It also gives higher water yield and has little effect on water quality (Zhang et al., 2016). Oxidants such as ozone, chlorine, and chloramine are usually used in drinking water treatment and can mitigate membrane fouling. However, various problems can occur during actual operation. Ozone always requires extra equipment, and chlorine and chloramine both produce disinfection by-products, such as trihalomethanes (THMs) and haloacetic acids (HAAs) (Tian et al., 2013; Jeong et al., 2014; Xu et al., 2014). Compared with these oxidants, hydrogen peroxide (H_2O_2) has many advantages, including mild disinfection effect, limited membrane damage, and safe decomposition by-products (Alasri et al., 1993; Farahbakhsh et al., 2004; Linley et al., 2012). However, most previous studies have been focused on the combined effects of H_2O_2 and other oxidants, with few studies investigating H_2O_2 alone with regard to the coagulation-ultrafiltration process in drinking water treatment (Song et al., 2004; Stanford et al., 2011; Pramanik et al., 2016). Furthermore, the influence of H_2O_2 on microorganisms and EPS, which are the major contributors to biofouling, is not clear.

Herein, we used the traditional coagulation–ultrafiltration membrane process to investigate membrane biofouling after H_2O_2 oxidation. Because of the strong corrosiveness of Fe-based salts during long-term operation, aluminum sulfate was used as the coagulant in this study (Esih et al., 2005). The aim of this research was to provide better understanding of the membrane biofouling mitigation following H_2O_2 pre-oxidation treatment, particularly its influence on the growth rate of microorganisms, proportion of proteins and polysaccharides in EPS, composition of proteins, and contribution of bacteria to membrane fouling.

1. Materials and methods

1.1. Materials

All reagents used were of analytical grade, except when specified. Both $Al_2(SO_4)_3{\cdot}18H_2O$ and H_2O_2 were purchased from Sinopharm Chemical Reagent, Co., Ltd. (China). Deionized (DI) water was used for preparing stock solutions during the whole experiment. To simulate micro-polluted surface water, domestic sewage was mixed with tap water at a volume ratio of 1:50 (Yu et al., 2014).

1.2. Experimental setup

Fig. 1 shows the schematic diagram of the experimental setup. The concentrations of $Al_2(SO_4)_3{\cdot}18H_2O$ and H_2O_2 were 0.05 mmol/L and 0.5 mmol/L, respectively. The rapid mix speed was 300 rpm for 1 min, after which it was decreased to 100 r/min for 14 min. A polyvinylidene fluoride (PVDF) hollow fiber UF membrane module (Motianmo, China) with an average pore size of 30 nm was used.

The total surface area of the submerged membrane in the membrane tank was 0.025 m^2. The constant permeate flux was 20 L/m^2/hr. After 30 min filtration, a 1 min backwash (40 L/m^2/hr) with air blowing (100 L/hr) was operated. This operation was cycled. The hydraulic retention time (HRT) of the membrane tank was 0.5 hr. During filtration, the transmembrane pressure (TMP) was monitored to reflect the development of membrane fouling. The sludge was discharged every 3 days. The residuals of H_2O_2 were quenched by NaClO.

1.3. Flow cytometry measurements of bacteria in the membrane tank water

Water samples were filtered by a cell strainer (300 mesh) to exclude flocs, and then mixed with ethylenediaminetetraacetic acid (EDTA) to improve the permeability of the outer cell membrane for gram-negative bacteria (Berney et al., 2007).

SYBR Green I (Invitrogen, USA) was used to stain all cells, and propidium iodide (PI; Sigma, USA) was used to stain dead or damaged cells with compromised cell membranes (Cerca et al., 2011). The stained samples were incubated in the dark at room temperature for 25 min, and were then measured by a flow cytometer (FACSCalibur 4CLR, BD Bioscience, USA).

1.4. Characterization of cake layer after filtration

At the end of the experiment (day 60), membrane fibers (2 cm) were cut from the two membrane modules. These fibers were fixed with 3.0% glutaraldehyde in 0.1 mol/L phosphate buffer at pH 7.2 and dehydrated with a graded ethanol series (Dizge et al., 2011). The membrane surfaces were then analyzed using a Hitachi SU8000 scanning electron microscope (SEM; Hitachi, Japan).

The sludge on the membrane surface was stained by Concanavalin A conjugated to Texas red (ConA-Texas red; Molecular Probes, USA), fluorescein isothiocyanate (FITC; Sigma, USA), and 4′, 6-diamidino-2-phenylindole, (DAPI;

Please cite this article as: Wang, X., et al., The effects of hydrogen peroxide pre-oxidation on ultrafiltration membrane biofouling alleviation in drinking water treatment, J. Environ. Sci. (2018), https://doi.org/10.1016/j.jes.2018.01.020