Membrane fouling in ultrafiltration of natural water after pretreatment to different extents

Lu Ao[1,2], Wenjun Liu[1,], Lin Zhao[1], Xiaomao Wang[1]*

1. School of Environment, Tsinghua University, Beijing 100084, China. E–mail: zyzzalou@163.com
2. Department of National Defence Architecture Planning & Environmental Engineering, Logistic Engineering University, Chongqing 401311, China

ARTICLE INFO

ABSTRACT

The combined fouling during ultrafiltration (UF) of surface water pretreated to different extents was investigated to disclose the roles of polysaccharides, proteins, and inorganic particles in UF membrane fouling. Both reversible and irreversible fouling decreased with enhanced pretreatment (biologically active carbon (BAC) treatment and sand filtration). The sand filter effluent fouled the membrane very slowly. The UF membrane removed turbidity to less than 0.1 nephelometric turbidity unit (NTU), reduced polysaccharides by 25.4%–29.9%, but rejected few proteins. Both polysaccharides and inorganic particles were detected on the fouled membranes, but inorganic particles could be effectively removed by backwashing. The increase of turbidity in the sand filter effluent to 3.05 NTU did not significantly increase the fouling rate, but an increase in the turbidity in the BAC effluent to 6.11 NTU increased the fouling rate by more than 100%. The results demonstrated that the polysaccharide, not the protein, constituents of biopolymers were responsible for membrane fouling. Membrane fouling was closely associated with a small fraction of polysaccharides in the feed water. Inorganic particles exacerbated membrane fouling only when the concentration of fouling–inducing polysaccharides in the feed water was relatively high. The combined fouling was largely reversible, and polysaccharides were the predominant substances responsible for irreversible fouling.

© 2015 The Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences.
Published by Elsevier B.V.

Introduction

Ultrafiltration (UF) has been increasingly applied for drinking water production due to its consistently high performance in terms of particle and microorganism removal (Boudaud et al., 2012; Hagen, 1998; Xiao et al., 2012). Optimally, UF can be operated as a stand-alone process with no requirement for pretreatment operations. However, one major challenge facing UF is membrane fouling, which impairs its cost efficiency, in that membrane fouling can decrease filtration flux and increase operational cost and, most severely, result in system failure. An understanding of membrane fouling is therefore crucial for better design and operation of UF processes.

Membrane fouling is caused by the accumulation of any substance (*i.e.*, a foulant) on the membrane surface or in the membrane material; a foulant restricts water flowing across the membrane. The foulants in natural waters include inorganic particles, natural organic matter (NOM), microorganisms, mineral ions (*e.g.*, calcium), and mixtures of these substances due to mutual interactions (Buffle et al., 1998). Nevertheless, NOM was demonstrated to be primarily responsible for UF membrane fouling (Cho et al., 2000; Escobar et al.,

* *Corresponding author.* E-mail: wjliu@tsinghua.edu.cn (Wenjun Liu).

2005; Shao et al., 2011; Wang and Wang, 2006; Yamamura et al., 2007; Yuan and Zydney, 2000). In particular, the biopolymers or colloidal fractions of NOM were more closely associated with membrane fouling than other NOM fractions, such as humic substances (Carroll et al., 2000; Katsoufidou et al., 2008; Kennedy et al., 2008). Measurements using a liquid chromatograph-organic carbon detector (LC-OCD) demonstrated that biopolymers were easily retained by UF membranes (Halle et al., 2009; Huang et al., 2011; Kennedy et al., 2005; Subhi et al., 2012). The major components of biopolymers are polysaccharides and proteins, but the polysaccharide component tends to contribute more to membrane fouling. Analyses using Fourier transform infrared (FT-IR) spectroscopy showed that polysaccharides were abundant on the fouled membrane surface (Cho et al., 1998; Kimura et al., 2004). Many polysaccharides in water, sometimes aided by calcium ions, can form a gel-like structure, which usually has very low filterability. Calcium ions are usually abundant in most natural waters and exceed the quantity needed for gel formation. Nevertheless, membrane fouling by polysaccharides can also be aggravated in the presence of proteins and humic substances (Gray et al., 2011). Fluorescence excitation–emission matrix (EEM) measurements showed that the protein peaks had some correlation with the membrane fouling rate (Kimura et al., 2014). Peldszus et al. (2011) found that protein-like substances contributed to both reversible and irreversible fouling.

Recently, much attention has been paid to the combined fouling caused by NOM coexisting with inorganic particles. The inorganic particles that are naturally present in or intentionally added to water include silts, clays, and precipitated crystals, as well as iron and aluminum oxyhydroxides (Yiantsios and Karabelas, 1998). Although inorganic particles alone usually do not lead to severe membrane fouling (Tian et al., 2013) and whatever fouling does occur is mostly reversible and easily removed by physical cleaning (Li et al., 2010; Peiris et al., 2010), previous studies showed that inorganic particles could aggravate membrane fouling caused by NOM. For example, membrane fouling by NOM "model" compounds (such as bovine serum albumin, humic acid and sodium alginate) increased in the presence of silica particles (Munla et al., 2012). Tian et al. (2013) also found that the membrane fouling rate was proportional to the concentration of silica particles in two size ranges (0.5–10 μm and 45 μm) in the range of 10–50 mg/L. However, a few studies showed that the coexistence of inorganic particles with NOM alleviated membrane fouling. Notwithstanding the above efforts, little is known about the combined fouling of membranes by NOM and inorganic particles found in natural water.

Pretreatment of water prior to filtration, usually using conventional drinking water treatment processes, has been proposed as a means to control membrane fouling. Coagulation was found to be effective in reducing membrane fouling (Chen et al., 2007; Citulski et al., 2008; Song et al., 2008) in that NOM could be substantially adsorbed to flocs of hydrolyzed coagulants that are highly filterable. Coagulation in combination with sedimentation was more effective than coagulation alone in reducing membrane fouling (Liang et al., 2008), because the pretreatment could effectively remove both NOM and inorganic particles from the water (Liu et al., 2011). Sand filtration was added following coagulation to control

membrane fouling, but the additional benefit was found to be minimal (Xia et al., 2004). Ozonation reduced membrane fouling because of its effectiveness in degrading biopolymers to small molecules that would be less likely to foul the membrane (Geismar et al., 2012; Guo et al., 2013, 2014). Biologically active carbon (BAC) filtration was shown to effectively decrease membrane fouling due to the retention of biopolymers in the filter (Guo et al., 2013; Halle et al., 2009; Huang et al., 2011).

In the study reported here, we investigated the combined fouling of UF membranes by NOM and inorganic particles found in natural water. The effects of various pretreatment techniques were examined, and the relative importance of polysaccharides and proteins in UF membrane fouling was evaluated. The water samples used as feed to the UF membrane were collected from a full-scale drinking water treatment plant (DWTP) at various stages in the treatment process. The full treatment process consisted of coagulation, followed by sedimentation, ozonation, up-flow BAC (UBAC) degradation, sand filtration, and chlorination (disinfection). The UBAC functioned more as a biological reactor than as a conventional down-flow BAC filter (Han et al., 2013). It was expected that both biopolymers and inorganic particles would be gradually removed throughout the treatment process. Special attention was paid to the differences in fouling potential of different polysaccharides in the feed water. A previous study showed that the polysaccharides could be roughly grouped into gelling and non-gelling components (Wang and Waite, 2009).

1. Materials and methods

1.1. Feed water

The feed water for the UF membrane experiments was from grab samples collected from three stages of treatment in the DWTP: after sedimentation (denoted as "FW-S" hereafter); after UBAC treatment (denoted as "FW-B"); and after sand filtration (denoted as "FW-F"). The dissolved oxygen (DO), conductivity, and pH of samples were measured on site. Thereafter, all samples were transported to the laboratory within 24 hr, where other water quality parameters including dissolved organic carbon (DOC), spectral absorption (UV$_{254}$), polysaccharide concentration, and turbidity were measured immediately upon arrival. The feed water samples were then stored in a refrigerator at 4°C and warmed to room temperature prior to use in the ultrafiltration experiments.

1.2. Ultrafiltration procedure

A laboratory-scale membrane filtration apparatus was used for all filtration experiments. The apparatus consisted of a home-made tubular filtration cell with an effective volume of 500 mL, two peristaltic pumps (BP100-1L, Longer, Hebei, China) to add feed water and provide permeate suction, a pressure transducer (CX-203, Futexin, Beijing, China) to measure trans-membrane pressure (ΔP), and other accessories (Fig. 1). A hollow fiber UF membrane module (Litree, Hainan, China) with an effective filtration area of 0.04 m^2 was installed in the tubular filtration cell. The membrane material was polyvinyl chloride and the nominal membrane pore size was 0.01 μm.