



Effect of pre-ozonation on mitigation of ceramic UF membrane fouling caused by algal extracellular organic matters



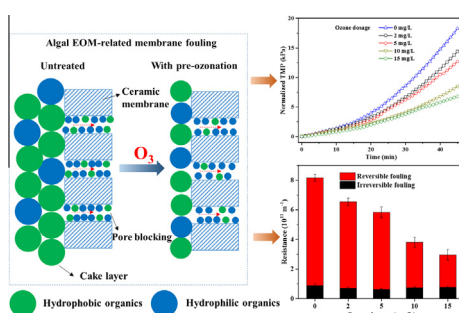
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HIGHLIGHTS

- Combined cake-standard model best fits the EOM-related fouling control performance.
- Standard pore blocking plays a more important role for the combined model.
- Pre-ozonation is highly effective to mitigate the EOM-related membrane fouling.
- The fouling mitigation is mostly attributed to the alleviation of reversible fouling.
- Ozone can selectively oxidize the very high MW hydrophobic biopolymers (≥ 20 kDa).

GRAPHICAL ABSTRACT



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ABSTRACT

Soluble extracellular organic matters (EOM) resulting from algal blooms in water sources can cause severe membrane fouling in water treatment. The effect of pre-ozonation on mitigation of ceramic UF membrane fouling caused by EOM released from *Microcystis aeruginosa* and the associated fouling mitigation mechanism were investigated through the characterization of hydraulic performance, dissolved organic carbon (DOC), fluorescence excitation–emission matrix (EEM) spectra, molecular weight (MW) distribution, hydrophilicity and model fit of five combined fouling models. Pre-ozonation achieved a remarkable effect of hydraulically reversible fouling mitigation as a result of the selective oxidation of the very high MW hydrophobic biopolymers (≥ 20 kDa) to lower MW and more hydrophilic compounds. However, pre-ozonation had a very limited effect on mitigation of hydraulically irreversible fouling dominated by the high MW (1–10 kDa) hydrophilic organics. Modeling results indicated that the EOM-related membrane fouling mitigation by pre-ozonation was more likely ascribed to the alleviation of cake layer and standard pore blocking with standard pore blocking playing a more important role.

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1. Introduction

The frequent occurrence of algal blooms in many lakes and reservoirs has brought a serious challenge to conventional drinking water treatment processes [1]. Algal blooms can not only intervene in the treatment processes but also deteriorate the water quality

with the release of a large amount of algal extracellular organic matters (EOM) [2].

Ultrafiltration (UF) membrane has been suggested as an ideal candidate for algal-rich water treatment with complete removal of algal cells and less release of intracellular toxins or odorous compounds [3,4]. However, membrane fouling remains a major obstacle for UF membrane application in algal-rich water treatment [3,4]. EOM have been recognized as the major membrane foulants to cause severe membrane fouling for both polymeric [2,5] and ceramic membranes [6,7] in algal EOM-rich water treatment. The very high molecular weight (MW) biopolymers (≥ 20 kDa), such as polysaccharides and proteinaceous substances, were proposed as the main EOM components for membrane fouling [6,8].

In order to mitigate the EOM-related membrane fouling, many pretreatment processes have been applied for the treatment of EOM-containing water, such as coagulation by ferric chloride [9], adsorption by mesoporous adsorbent resin [10], and preoxidation by UV/H₂O₂ [11]. Ozone, a powerful oxidant with a redox potential of 2.07 V, has been widely used in drinking water treatment [12]. Moreover, ozonation has been demonstrated to effectively reduce membrane fouling caused by natural organic matters (NOM) and secondary effluent [13,14]. Molecular ozone and resultant hydroxyl radicals can oxidize NOM and change their hydrophilicity and polarity with the resultant mitigation of membrane fouling [15]. Furthermore, ozonation with low ozone dosages (<1.0 mg O₃/mg DOC) can break down high MW NOM to lower MW molecules [16,17]. Comparing to polymeric membranes, ceramic membrane has robust mechanical and chemical stability, long life span and high hydrophilicity and it can withstand the severe oxidizing conditions with the presence of ozone and hydroxyl radicals [18]. In recent years, an integrated process of ozonation and ceramic membrane filtration has been proposed for effective fouling mitigation [19–22]. As such, the integrated application of ozonation and ceramic UF membrane filtration could be an alternatively feasible pathway for the treatment of algal EOM-rich water with the effective removal of algal EOM and mitigation of membrane fouling.

In-situ ozonation coupled with ceramic UF membrane filtration was very recently proposed for the treatment of algal-rich water with effective removal of algal cells and mitigation of membrane fouling [22]. As for EOM-rich water, however, how ozonation facilitates the mitigation of algal EOM-related membrane fouling from the perspective of cake layer, gel layer and membrane pore blocking remains unclear.

In this study, we applied an integrated process of pre-ozonation and ceramic UF membrane filtration for algal EOM-rich water treatment with objectives to 1) demonstrate the effectiveness of pre-ozonation on mitigation of ceramic UF membrane fouling caused by EOM released from *Microcystis aeruginosa* at lab scale, and to 2) explore the associated fouling mitigation mechanism with five combined fouling models.

2. Materials and methods

2.1. Algal EOM solution preparation

M. aeruginosa (FACHB 905) with the spherical shape and cell size of 2–7 μm was purchased from Freshwater Algae Culture Collection of the Institute of Hydrobiology (FACHB Collection, Wuhan, China). The algal cultures were grown in BG-11 medium (Table S1) in a 10 L narrow-mouthed bottle placed in an incubation chamber (Keelrein, China) at 25 °C with aeration. Fluorescent lamps (Philips, China) were used as the light source with an automated light/dark cycle of every 12 h and light intensity of 2000 lx during the lighting phase. Cultures were harvested at the

stationary phase after a 30-day incubation with the cell density of 2.3×10^7 cells/mL.

Algal EOM solution was extracted by two steps [3]: 1) centrifugation of the harvested algal cultures at 10,000g and 4 °C for 15 min (5804R, Eppendorf, Germany); and 2) subsequent filtration of the supernatant by the mixed cellulose ether membrane with 0.45 μm pore size (Jinteng, China). The DOC content of EOM solution was diluted to 10.5 ± 0.5 mg/L with Milli-Q water for severe membrane fouling study as well as membrane fouling control study by pre-ozonation.

2.2. Pre-ozonation treatment

The pre-ozonation set-up is shown in Fig. S1. Ozone was generated by an ozone generator (OF-G-3-10g, QingDao Guolin, China). The ultra-pure oxygen from a gas cylinder went through a silica gel drier and was fed to the ozone generator. The concentration of gaseous ozone was adjusted by varying the current of the generator and the concentration was measured by an ozone monitor (IDEAL-2000, Aidier, China). The gas flow containing ozone was injected directly at the bottom of the reactor flask by a titanium aeration rod. The excess ozone after ozone monitor and the non-reacted ozone at the outlet gas of the vessel were passed into the 2% KI solution to get rid of any residual ozone. Pre-ozonation of 2.0 L EOM solution with different gaseous ozone concentrations of 0, 2, 5, 10 and 15 mg/L was conducted before the filtration tests. The pH of the EOM solutions before and after the treatment was adjusted to 7 using 1 mol/L HCl or NaOH.

2.3. UF filtration tests

The set-up of ceramic UF membrane experimental filtration is shown in Fig. 1. The membrane tank is a cubic plexiglass container with the inner dimension of 150 mm (length) \times 60 mm (width) \times 300 mm (height). A flat sheet Al₂O₃ ceramic membrane (Meidensha Corporation, Japan) with an average pore size of 60 nm and the porosity of 42.1% and the dimension of 80 mm (length) \times 70 mm (width) \times 6 mm (thickness) was vertically fixed in the membrane tank. The membrane surface and cross-section morphologies were shown in Fig. S2. The water in the membrane tank was maintained at a constant level by two peristaltic pumps (Longer Pump, China), a feed pump and a sucking pump, operated at the same rotation rate. A pressure gauge (GC31-174, Nagano Keiki, Japan) was mounted between the membrane tank and the suction pump to monitor the trans-membrane pressure (TMP). A flowmeter (VN05, Aichi Tokei Denki, Japan) was mounted between the suction pump and the permeate tank to monitor the flow. The pressure gauge and flowmeter were connected to a computer, and the data acquisition system automatically recorded the pressure data (referred as TMP data) and the flowmeter data every one minute. Normalized TMP was calculated as the difference between the final TMP and the initial baseline. All filtration tests were run in duplicate.

1.2 L EOM solution from the pre-ozonation treated 2.0 L solution was poured into the membrane tank before the filtration, and the rest 0.8 L solution was then fed into the membrane tank by the feed pump during the filtration. A same protocol with a filtration flux of 90 L/m²·h and a running period of 45 min was applied for all experiments. After each UF test, the fouled membrane was cleaned following two steps: 1) backwash with 150 mL Milli-Q water (physical cleaning); and 2) backwash with 150 mL HCl solution (0.01 mol/L) followed by 150 mL NaOH solution (0.01 mol/L) and then soaking in NaClO solution (100 ppm available chlorine) (chemical cleaning). The same ceramic membrane was used for all UF filtration tests.

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