



A comparative study of pre-ozonation and *in-situ* ozonation on mitigation of ceramic UF membrane fouling caused by alginate



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ABSTRACT

Pre-ozonation or *in-situ* ozonation coupled with membrane filtration has been actively utilized for membrane fouling control in water treatment, however, the comparative analysis of these two ozonation processes on membrane fouling control has not been reported so far. In this study, the comparative effect of pre-ozonation and *in-situ* ozonation on mitigation of ceramic UF (ultrafiltration) membrane fouling caused by alginate was systematically investigated through characterization of TMP, fouling resistance, MW distribution, dissolved ozone concentration, the formation of hydroxyl radical and modeling. Results showed that both pre-ozonation and *in-situ* ozonation were effective in alleviating the cake layer fouling. Ozonation efficiently broke down the high molecular weight (MW) fractions (> 150 kDa and 80–150 kDa) of alginate into the low MW fractions (6–80 kDa; 0.4–6 kDa and 0.1–0.4 kDa), reducing the membrane interception of alginate. Pre-ozonation achieved better fouling mitigation performance compared with *in-situ* ozonation at low ozone dosages of 2 and 4 mg/L. However, *in-situ* ozonation had better fouling control performance at high ozone dosage of 10 mg/L with the normalized TMP (Δ TMP) maintaining below 0.2 kPa after 60 min filtration. Besides, *in-situ* ozonation was more effective in hydraulically irreversible fouling control than pre-ozonation regardless of ozone dosage. Integration of *in-situ* ozonation and ceramic membrane filtration catalyzed ozone decomposition with the increased generation of hydroxyl radical, which further strengthened the oxidation of the accumulated foulants on the membrane surface and within the membrane pores and thus effectively alleviated the reversible and irreversible fouling.

1. Introduction

The implementation of membrane process nowadays is considered as a well-established technology for the treatment of contaminated natural waters with the advantages of small footprint and high water quality of permeate. Recently, ceramic membranes have been actively utilized in water treatment with the merits of ceramic membranes of long life span, high hydrophilicity, robust thermal, chemical and mechanical stability, high permeate flux at low transmembrane pressure [1–3]. However, membrane fouling is still a big issue in the applications of ceramic membranes as membrane fouling can lead to the decline of flux and increment of energy and chemical consumption [4]. Natural organic matters (NOM) are the major foulants of membrane fouling in the treatment of natural waters [5–10]. Meanwhile, the hydrophobicity and molecular weight (MW) of NOM also significantly

affect membrane fouling [5]. For instance, the hydrophilic fraction of NOM was suggested to contribute greatest to membrane fouling [7] and the very high MW biopolymers (> 20 kDa) were proposed as the main components depositing on the membrane surface to form a thick and dense membrane fouling layer [11].

Pre-ozonation has been used to mitigate membrane fouling as ozone can oxidize NOM to lower MW substances [12] and change the molecular structure [10,13] and hydrophilic proportion of NOM [14]. Such changes primarily resulted from the preferential oxidation of electron-rich moieties containing carbon-carbon double bonds and aromatic rings [12,15] in humic acid (HA) [15], proteins [16], and polysaccharides [17]. Bae observed that pre-ozonation can slow down the increment of transmembrane pressure (TMP) during the ultrafiltration (UF) membrane filtration process [18]. Geno Lehman and Liu also found that pre-ozonation was effective on the degradation of colloidal NOM [3].

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Nomenclature

EOM	extracellular organic matters
HA	humic acid
MW	molecular weight
NOM	nature organic matters
SA	salicylic acid

SA-3	2,3-dihydroxy benzoic acid
SSE	sum of squares of difference between experimental data and fitted values
TMP	transmembrane pressure
UF	ultrafiltration
Δ TMP	normalized TMP increase

However, pre-ozonation could not alleviate the irreversible fouling including the gel layer formation on the membrane surface and pore blocking in the membrane pores formed during the filtration process [19]. The medium and low MW fractions of organics mainly contribute to the irreversible fouling, for which chemical cleaning is required [19].

Ceramic membrane-based catalytic ozonation process has recently been gained significant attention as a more effective treatment process for mitigation of membrane fouling, degradation and mineralization of organic pollutants in water treatment [9,12,20,21]. The fouling control mechanisms of the integrated process of *in-situ* ozonation and ceramic membrane filtration include molecular ozone directly oxidation and hydroxyl radical oxidation of feed and foulants deposited on the membrane surface and within the membrane pores [4,22]. In our previous studies, both reversible and irreversible fouling were effectively mitigated by *in-situ* ozonation in algal-rich water [23], wastewater [24] and drinking water treatment [25].

However, whether *in-situ* ozonation outweighs pre-ozonation in view of membrane fouling control efficiency remains unclear even though *in-situ* ozonation is effective in controlling membrane irreversible fouling. For example, *in-situ* ozonation starts simultaneously with ceramic membrane filtration of natural waters and the feed usually has higher MW fractions of NOM compared with the feed by pre-ozonation in which the high MW fractions of NOM have already been oxidized to lower MW substances before filtration. Therefore, *in-situ* ozonation might have more severe reversible fouling caused by the high MW fractions of NOM compared with pre-ozonation [9,12].

We previously studied the respective effect of pre-ozonation and *in-situ* ozonation on ceramic membrane fouling control in algal-rich water [23,26] and wastewater treatment [24,27]. Our results indicated that pre-ozonation might achieve the similar TMP decrement as *in-situ* ozonation did even though the fouling mitigation mechanism for pre-ozonation was different from that for *in-situ* ozonation. Pre-ozonation remarkably reduced organics in feed before filtration, alleviating the reversible fouling formed during the filtration process [26,27]. *In-situ* ozonation reduced organics in feed and on the membrane surface and/or in the membrane pores, alleviating both the reversible and irreversible fouling formed during the filtration process [23,24]. Many previous studies also reported the effect of pre-ozonation [28–30] and *in-situ* ozonation [9,22,31,32] on membrane fouling control with only focusing on one side (either pre-ozonation or *in-situ* ozonation), however, the comparative analysis of these two ozonation processes on membrane fouling control has not been reported so far.

Moreover, more detailed explanations of the fouling mitigation mechanisms of organic induced membrane fouling by pre-ozonation and/or *in-situ* ozonation are required to better understand the fouling transformation in ceramic membrane-based water treatment processes. In this study, alginate was chosen to represent polysaccharide in natural waters. The main objective of this work was to study systematically the influence of ozone dosage (0, 2, 4, 10 mg/L) on membrane fouling control with pre-ozonation and *in-situ* ozonation. The fouling mitigation mechanisms were explored through characterization of TMP, fouling resistance, MW distribution, dissolved ozone concentration, the formation of hydroxyl radical and modeling.

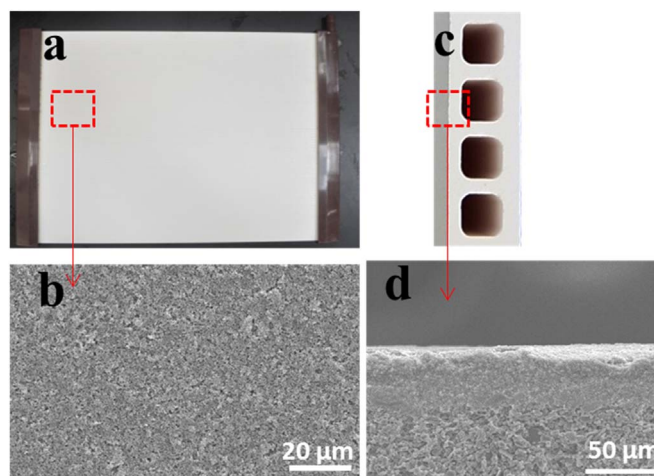


Fig. 1. Photographs of ceramic UF membrane surface (a) and cross-section (c), SEM micrographs of ceramic membrane surface (b) and cross-section (d).

2. Materials and methods

2.1. Ceramic membrane

Flat sheet Al_2O_3 ceramic ultrafiltration (UF) membrane (Meidensha Co., Japan) with a total filtration area of 0.042 m^2 (250 mm (length) \times 80 mm (width) \times 6 mm (thickness)) and the mean pore size of 100 nm is used and its structure is shown in Fig. 1.

2.2. Experimental design

The schematic diagram of the hybrid process of ceramic membrane filtration and ozonation is shown in Fig. 2. The cubic plexiglass membrane tank (inner dimension of 500 mm (length) \times 150 mm (width) \times 50 mm (thickness)) was fixed on a magnetic stirring

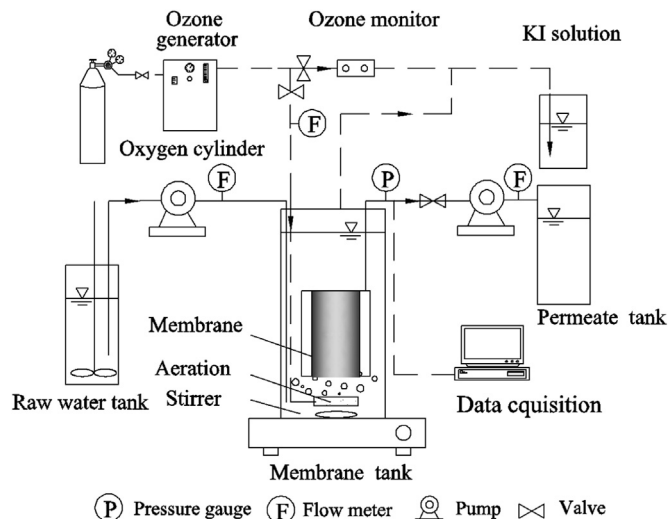


Fig. 2. Schematic diagram of ceramic membrane filtration experiment with ozonation.

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