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Pretreatment strategies for mitigation of membrane fouling by effluent organic matter in ultrafiltration: ozonation and coagulation/flocculation

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ABSTRACT

Membrane separation processes are widely used in wastewater reuse systems. However, membrane fouling is a challenging issue because it leads to high energy consumption and decreases separation efficiency. In this study, we investigated measures to control the membrane fouling arising from the deposition of foulants such as particles, colloids, and effluent organic matter onto the membrane surface during secondary wastewater management. Preozonation and coagulation/flocculation pretreatments were tested as measures to reduce membrane fouling. Filtration resistance was notably high in the absence of preozonation and coagulation/flocculation pretreatments; in contrast, both preozonation and coagulation/flocculation facilitated the removal of organic foulants by decreasing the irreversible fouling during filtration. The optimal ozone contact time for preozonation was found to be 120 s, in terms of improving the permeate water flux. In addition, the use of iron(III) chloride ($FeCl_3$; dosage 25 mg Fe/L) with preozonation was required to depolarize foulants from the membrane surface. These findings suggest that irreversible resistance can be controlled by using preozonation and coagulation/flocculation pretreatments after eliminating the reversible phase. Therefore, the combined application of ozone and coagulant can play a key role in stable operation of the membrane separation process for wastewater reuse.

Keywords: Effluent organic matter; Membrane; Fouling; Ozonation; Coagulation

1. Introduction

Water scarcity is a growing problem that is expected to affect 60% of the global population in 2025. There is a great need for clean and stable water supplies [1–3]. Urban wastewater has recently been evaluated as an alternative water source to decrease water scarcity. As a water source, reclaimed wastewater that has undergone conventional treatment in a wastewater treatment plant (WWTP) has the advantages of minimizing changes in the quantity and quality of water, as well as stability of supply [4]. It is widely reused for replenishing surface reservoirs and groundwater basins, and in water-scarce areas, it is often used for landscaping and agriculture [5].

Recently, water reclamation methods such as indirect potable reuse and direct potable reuse have been studied to enhance public acceptance [6]. The development of filtration technologies has provided practical means of achieving high removal efficiency at a reasonable cost. Membrane filtration technologies can be divided into microfiltration (MF), ultrafiltration (UF), nanofiltration, and reverse osmosis (RO). Brackish water reverse osmosis (BWRO), which is capable of removing effluent organic matter

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(E_fOM), pesticides, pharmaceuticals, endocrine disruptors, salts, and other inorganic species can be applied to wastewater reuse [7]. In addition, low-pressure MF and UF processes can be applied as pretreatments prior to BWRO to prevent fouling by suspended solids, particles, viruses, and colloids [8–12]. MF and UF can be effective in removing the particulate E_fOM, large colloids, and bacterial cells from wastewater [13]. However, the fouling caused by E_fOM, colloids, and particles capable of forming fouling layers when deposited on the membrane surface remains the biggest problem [14]. The organic composition of E_fOM in reclaimed wastewater cannot be uniquely identified, and there have been difficulties in providing a unified definition of E_fOM due to its high structural and functional complexity [15].

A recent approach based on fluorescence excitation and emission spectroscopy demonstrated that tyrosine-like and tryptophan-like substances as well as fulvic-like and humic-like substances were the main constituents of E_fOM [16,17]. It is desirable to apply pretreatment processes such as advanced oxidation, coagulation, and adsorption to avoid membrane fouling because the fouling not only increases electricity costs but also affects filtration performance [18–20]. Among these processes, oxidation (e.g., ozone treatment) has been studied over the past 30 years and is used for various water reclamation applications including the destruction of toxic organics such as anthropogenic and synthetic organic materials [21–23]. The advantages of oxidation are the ability to generate high concentrations of the hydroxyl radical, which is a strong oxidant capable of completely oxidizing most organic compounds and killing resistant pathogens. Ozone degrades to form hydroxyl radicals in water, and thus can react with functional groups such as the double bonds and aromatic rings of organic molecules [24,25]. Ozone oxidation does not remove dissolved organic carbon (DOC), but rapidly reduces its aromaticity as represented by specific UV absorbance (SUVA) or UV absorbance at 254 nm (UVA_{254}). The oxidation of E_fOM by ozone has high potential to improve membrane flux in the case of organic fouling [23,26,27].

Coagulation can reduce the membrane fouling that arises during filtration due to the aggregation of fine particles caused by the pore-blocking cake layer. The incorporation of such fine particles into highly porous flocs prevents pore blockage [28]. Flocculation is effective in removing compounds of both low and high molecular weight [29]. However, the flocs that are formed by coagulation can be damaged due to the properties of flocs [30]. The reduction of E_fOM molecular weight by means of ozone oxidation can improve coagulation and water quality [31–34]. Therefore, in the present work we examined the differences in treated water quality and UF membrane filtration resistance arising from the use of various conditions of ozonation and coagulation pretreatment. To predict the effects of preozonation (PO), fluorescence excitation emission matrix (FEEM), and Fourier transform infrared spectroscopy (FTIR) analyses were utilized to establish correlations with the conditions of ozonated water. Moreover, an online intelligent photometric dispersion analyzer (iPDA) was used to measure floc size value (FSV) during the coagulation/flocculation (CF) process.

2. Materials and methods

2.1. Source water

Secondary effluent samples for the experiments were obtained from the WWTP in Suwon, Republic of Korea; the water collected had been treated by means of conventional activated sludge treatment. Before laboratory testing, particles in the wastewater samples were removed using a 100 μm auto-strainer; the resulting filtered water was used as feedwater for the experiments. Table 1 lists characteristics of the feedwater. The parameters analyzed were turbidity, color, DOC, UVA_{254}, SUVA, chemical oxygen demand (COD), total nitrogen (T-N), total phosphate (T-P), total dissolved solids (TDS), and pH.

2.2. Ozonation

The PO experiments were carried out using a filtration system, which was used to perform both a flux decline rate (FDR) test and a bench-scale test with an ozone production of 2.5 ± 0.02 g/h. Ozone contact time of 30, 60, 90, and 120 s were used to predict the effects of PO; ozone was generated by an ozonizer (LAB-1, Ozonetech, Republic of Korea) equipped with a pure oxygen supply (99%). Ozone gas was injected at 25°C into the bottom of the ozone reactor using a bubble stone aerator.

2.3. Coagulation and flocculation

CF treatment with iron(III) chloride ($FeCl_3$) coagulant (Samchun, Republic of Korea) was applied to enhance the flux by removing the E_fOM in the secondary effluent. CF was conducted in a jar tester (C-JT, Chang Shin Scientific Co., Republic of Korea); various concentrations of $FeCl_3$ between 0 and 25 mg Fe/L were applied. In these experiments, the agitation speeds of the rapid mixing and slow mixing were 250 and 40 rpm, respectively, and the stir times were 1 and 15 min, respectively. For FDR tests, coagulants were added under agitation of 250 rpm, which was applied for 1 min.

2.4. Dead-end filtration tests for optimal doses of ozone and coagulants

Dead-end filtration tests were conducted to determine the optimal pretreatment doses of ozone and $FeCl_3$. The experimental setup of the filtration cell (Amicon 8200, Millipore, USA) and dispensing pressure vessel used for the

Table 1
Characteristics of feedwater

Parameters	Value
Turbidity (NTU)	3.35 ± 0.02
Color (Pt–Co)	72 ± 1.53
DOC (mg/L)	6.22 ± 0.08
UVA_{254} (cm^{-1})	0.112 ± 0.005
SUVA (m^{-1} mg^{-1} L)	1.80 ± 0.06
COD (mg/L)	26 ± 1.53
T-N (mg/L)	8.6 ± 0.15
T-P (mg/L)	0.63 ± 0.03
TDS (mg/L)	231 ± 1.53
pH	7.53 ± 0.04

FDR tests consisted of a flat sheet polyvinylidene fluoride (PVDF) membrane (GVWP04700, Millipore, USA) with an effective area of 28.7 cm^2. A membrane of pore size 0.22 μm, porosity 70%, and diameter 63.5 mm was fixed in the filtration cell. The nitrogen gas for filtration was connected to the dispensing pressure vessel and supplied at a constant pressure of 0.05 MPa. Before the FDR experiment, the initial membrane flux was measured using pure water at 0–0.1 MPa (0–3.862 ± 95.6 L/m^2/h). For each experiment, the permeate flux was monitored for 1,000 s using an electronic balance, and recorded by an acquisition system.

2.5. Bench-scale filtration tests

The bench-scale system setup for measurement of reversible and irreversible resistance applied by hydraulic backwash consisted of an auto-strainer (100 μm), PO, CF, and a filtration system. PVDF hollow fiber modules (HFU-LAB0.18, Toray Industries, Inc., Japan) were used for filtration. The molecular weight cutoff (MWCO) of the membrane was 150 kDa. The filtration system consisted of a UF module with an effective area of 0.18 m^2. All experiments were performed in dead-end mode under a constant flux of 60 LMH. During filtration, the filtration and hydraulic backwashing times of 29.5 and 0.5 min, respectively, were applied to periodically depolarize foulants from the membrane; five of these 30-min cycles were applied in each experiment. For the CF experiment, a line mixer was used for rapid mixing.

2.6. Membrane fouling analysis

To investigate the effects of PO and CF upon membrane fouling behavior, the total resistance of irreversible and reversible fouling was calculated from the flux and transmembrane pressure (TMP) using Darcy's equation as follows:

$$R_{\text{total}} = \frac{\Delta P}{\mu J} = R_{\text{membrane}} + R_{\text{fouling}} \tag{1}$$

where R_{total} is the total membrane resistance (m^{-1}), μ is assumed to be the dynamic viscosity of water (Pa s), J is the permeate water flux (L/m^2/h), ΔP is the TMP (Pa), R_{membrane} is the intrinsic membrane resistance, and R_{fouling} is the net hydraulic resistance (m^{-1}). R_{fouling}, the net hydraulic resistance due to fouling, is defined as the difference between R_{total} and R_{membrane}, as shown in Eq. (4). R_{membrane} can be obtained according to Eq. (2) by means of a pure water flux test, in which case R_{fouling} is zero. After filtration using secondary effluent samples, R_{total} was calculated according to Eq. (3):

$$R_{\text{membrane}} = \frac{\Delta P_0}{\mu J} \tag{2}$$

$$R_{\text{total}} = \frac{\Delta P_1}{\mu J} \tag{3}$$

Then, R_{fouling} was calculated according to Eq. (4). R_{fouling} includes the effects of $R_{\text{reversible}}$ and $R_{\text{irreversible}}$, which are the hydraulic reversible resistance (m^{-1}) and irreversible resistance (m^{-1}), respectively. After the filtration process, the TMP before physical cleaning of the final contaminated membrane was determined to be the reversible membrane fouling, and the magnitude of TMP not recovered after physical cleaning was considered to be the irreversible membrane fouling (ΔP_2). The values of $R_{\text{reversible}}$ and $R_{\text{irreversible}}$ were calculated by means of Eqs. (5) and (6).

$$R_{\text{fouling}} = \frac{\Delta P_1}{\mu J} - \frac{\Delta P_0}{\mu J} \tag{4}$$

$$R_{\text{reversible}} = R_{\text{fouling}} - \frac{\Delta P_2}{\mu J} \tag{5}$$

$$R_{\text{irreversible}} = R_{\text{total}} - R_{\text{membrane}} - R_{\text{reversible}} = \frac{\Delta P_2}{\mu J} \tag{6}$$

2.7. Analytical methods

The ozone generated in contact with ultrapure water was measured by means of the indigo method, using a UV–Vis recording spectrometer (DR6000, Hach, USA). Turbidity in nephelometric turbidity units was measured by using a turbidimeter (2100AN, Hach, USA). T-N, T-P, and COD were measured by using a vial kit provided by Hach Co., and color was measured by using the Pt–Co method. DOC was measured using a total organic carbon analyzer (TOC-LCPN, Shimadzu, Japan). Absorbance at 254 nm was measured using a UV–Vis spectrometer (DR6000, Hach, USA). TDS and pH were measured using a multimeter (HQd portable meter, Hach, USA). To monitor the floc size during CF, the FSV was measured using an online iPDA (iPDA-100, Econovel, Republic of Korea). FTIR analysis was conducted using a Tensor 27 IFS-66/S instrument (Bruker Optics). FEEM was measured using a spectrofluorometer (RF-6000, Shimadzu, Japan). Prior to analysis, all secondary effluent samples were filtered using a 100 μm auto-strainer.

3. Results and discussion

3.1. Ozonation efficiency

To determine the optimal ozone dose, we compared the removal efficiencies of turbidity, color, DOC, UVA$_{254}$, COD, T-N, and T-P arising from the use of contact time of 30, 60, 90, and 120 s. Removal of turbidity and T-P were apparent for contact time of 60 s, with removal efficiency maximized at 53.1%, and 17.5% for 120 s, respectively (Fig. 1). Color removal was apparent for all doses studied, but increased sharply between the contact time of 30 and 60 s; the maximum of approximately 72.2% was observed for 120 s.

With regards to organic matter, PO showed no removal of DOC, and removal of UVA$_{254}$ and COD gradually increased with increasing reaction time. Contact time of ozone at 120 s yielded UVA$_{254}$ and COD removal efficiencies of 56.0% and 31.8%, respectively. Due to the oxidation of organic matter, ozonation changed the FTIR spectra of E$_f$OM in the secondary effluent, with increases and decreases of the intensities in bands corresponding to various functional groups. FTIR spectra of the secondary effluent showed peaks at the wavelengths of 3,400 cm^{-1}, corresponding to O–H stretch of polysaccharides; 2,940 cm^{-1}, corresponding to aliphatic C–H stretch in humic substance; 1,640 cm^{-1}, corresponding to C=O bonds in primary amides within proteins; 1,400 cm^{-1}, corresponding to symmetric carboxylate stretch; and 1,170 cm^{-1}, corresponding to asymmetric CO–O–C stretch (Fig. 2). The polysaccharides and proteins evident from this analysis indicated the presence of humic substances and soluble microbial products (SMPs) [35]. PO treatment led to reduced intensity of the FTIR peaks associated with polysaccharides, proteins, and humic substances, which are capable of causing membrane fouling; also, the reduction in these peaks increased with increasing ozone dose. Thus, ozone injection was effective in the oxidation of organic matter.

FEEM spectra were acquired of untreated and ozone-treated secondary effluent samples (Fig. 3). These spectra can be divided into five regions as follows: regions I (excitation/emission: 220–250 nm/280–330 nm) and II (excitation/emission: 220–250 nm/330–380 nm) correspond to aromatic proteins, region III (excitation/emission: 220–250 nm/380–580 nm) corresponds to a fulvic-acid-like substance, and regions IV (excitation/emission: 250–470 nm/280–380 nm) and V (excitation/emission: 250–470 nm/380–580 nm) correspond to SMPs and humic-acid-like substances [36].

For untreated samples, the emission intensity was lower in the regions assigned to aromatic proteins and SMPs, whereas the regions assigned to humic-acid-like and fulvic-acid-like substances had comparable intensities (Fig. 3(a)). In samples treated by ozonation with contact time of 120 s, the intensities decreased by approximately 90.6% (II), 94.3% (III), 96.0% (IV), and 94.6% (V) relative to the intensities of the untreated samples (Fig. 3(b)). This indicated that ozonation readily removed proteins such as tyrosine-like and tryptophan-like substances, as well as fulvic-like and humic-like substances.

3.2. Coagulation/flocculation efficiency

3.2.1. Variations in water quality metrics

When applying filtration pretreatment prior to RO in the wastewater reuse process, the removal efficiency of particulate matter is perfect. However, it is not possible to

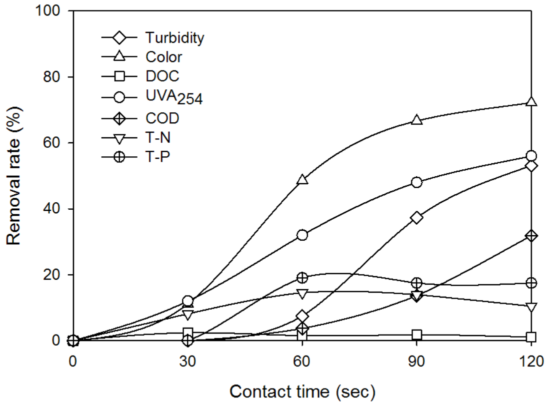

Fig. 1. Effects of ozonation on the removal of turbidity, color, DOC, UVA$_{254}$, COD, T-N, and T-P in the secondary effluent with increasing contact time.

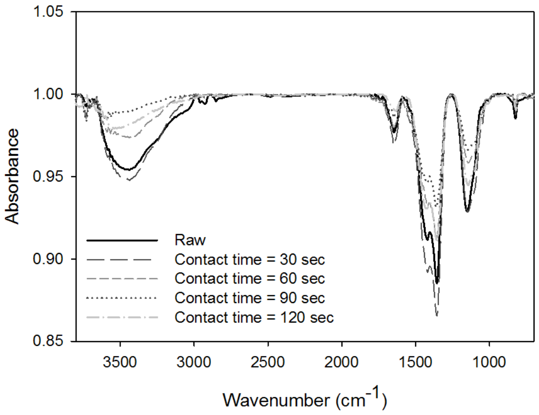

Fig. 2. Effects of ozonation on changes in FTIR spectra of E$_f$OM with increasing contact time.

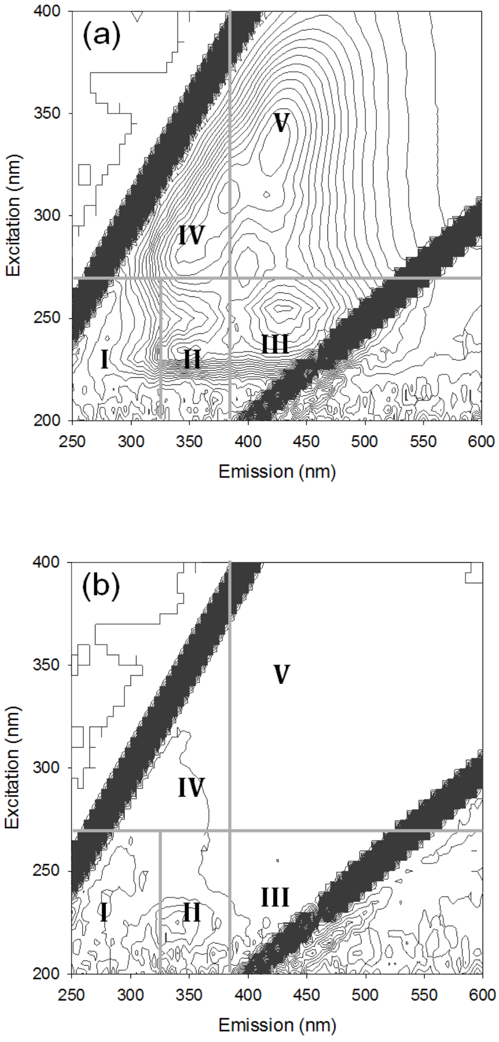

Fig. 3. Effects of ozonation on FEEM of E$_f$OM: (a) without ozonation and (b) with ozonation (contact time of 120 s).

expect perfect removal of E_fOM or trace organics with small molecular weights, or of ionic materials including heavy metals. Therefore, we attempted to determine the optimal coagulant dose to reduce the FDR of the membrane. Experiments were performed using injected doses of 5, 10, 15, 20, and 25 mg Fe/L. We analyzed the turbidity, DOC, and UVA_{254} of the supernatant 15 min after precipitation and then compared their removal efficiencies for each coagulant dose (Fig. 4). The turbidity, DOC and UVA_{254} removal efficiencies tended to increase with increasing $FeCl_3$ dose.

The DOC removal efficiency of approximately 56.3% was observed when the coagulant dose of 25 mg Fe/L was used, and the removal efficiency of UVA_{254} was around 42.9% for the dose of 25 mg/L. The highest turbidity removal efficiency of approximately 63.9% was observed when the 25 mg/L dose was used.

3.2.2. Changes in floc size

To examine the performance of CF when combined with PO treatment, we compared the behaviors of floc growth vs. treatment time for CF-only and combined PO/CF treatments (Fig. 5). Floc size in FSV was measured for samples undergoing treatment; this was carried out by using an iPDA system including a light source of wavelengths 550–900 nm. Prior to floc size measurement, each sample was rapidly agitated for 1 min in a jar tester. Then, the sample was slowly agitated and floc size was measured for approximately 300 s.

The FSV increased from 0.1 to 0.13 without injection of ozone and coagulant as a pretreatment. On the other hand, it increased to 0.18 with the injection of 25 mg/L $FeCl_3$. The FSV increased to approximately 0.44 with the injection of 25 mg/L $FeCl_3$ after ozone contact time of 120 s. Thus, the CF performance improved when ozone was injected prior to the injection of $FeCl_3$.

3.3. Evaluation of FDR

FDR tests were conducted with various ozone doses. The FDR was 63.7% without any ozone, and the FDR decreased to 61.3% and 57.9%, respectively, when contact time of 90 and 120 s were used (Fig. 6(a)). Thus, ozone treatments of these two doses increased the permeability. Contrastingly, the FDR was 63.8% and 64.7%, respectively, when the lower

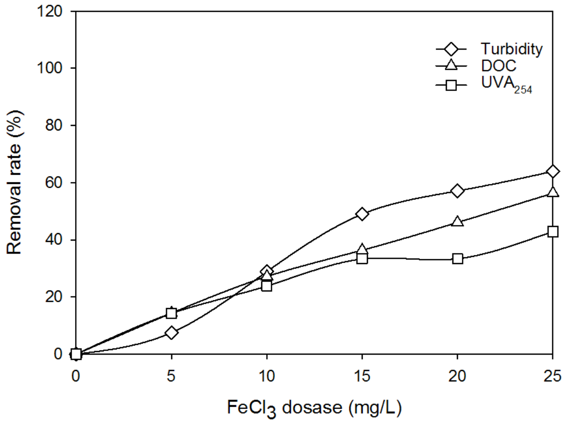

Fig. 4. Removal efficiency of turbidity, DOC, and UVA_{254} by coagulation/flocculation.

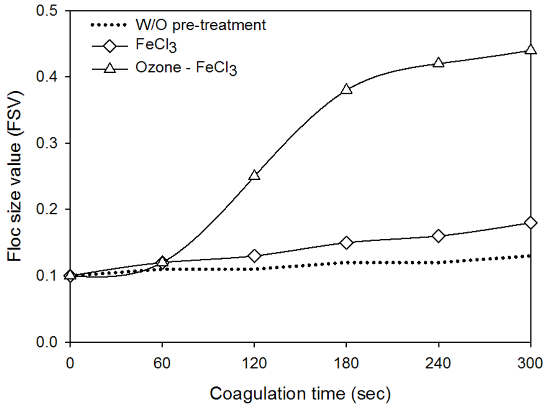

Fig. 5. Effects of pretreatment strategies using ozonation and coagulation/flocculation on the floc formation and floc size distribution.

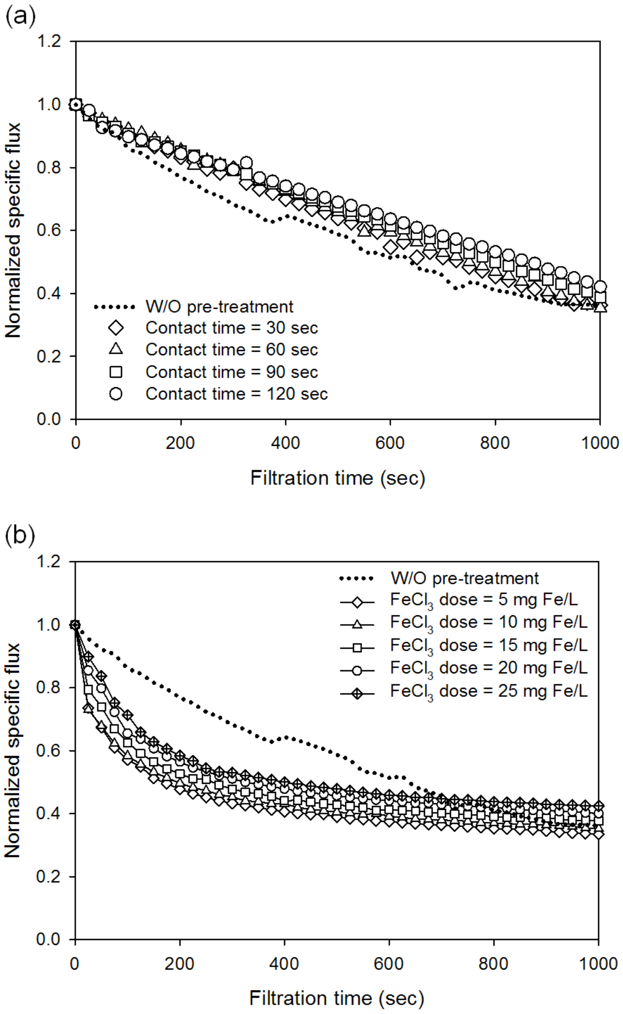

Fig. 6. Changes in FDR of the dead-end filtration tests: (a) ozonation and (b) coagulation/flocculation.

contact time of 30 and 60 s were used, representing worse permeability compared with the control case.

To evaluate the FDR in condition of FeCl$_3$ doses, FeCl$_3$ was injected at the concentrations of 5, 10, 15, 20, and 25 mg Fe/L, and the FDR was then measured. The FDR was the lowest, 57.5%, when the FeCl$_3$ dose of 25 mg Fe/L were used (Fig. 6(b)). Conversely, the FDR was 66.5% when the FeCl$_3$ dose of 5 mg Fe/L was used. Therefore, it was necessary to evaluate the appropriate doses of ozone and coagulant to minimize fouling. The combination of 120 s (ozone contact time) and 25 mg Fe/L (FeCl$_3$) showed the highest removal efficiency in this study.

3.4. Effects of PO and CF upon reversible and irreversible membrane fouling

Differences in the filtration resistance with and without the use of the PO, CF, and combined PO/CF pretreatments were evaluated using PVDF UF membranes with MWCO 150 kDa. Dead-end filtration was performed at 60 LMH, and physical backwashing was applied to evaluate the reversible and irreversible fouling resistances. Changes in the filtration resistance during approximately six cycles were measured after applying the flow rate of backwash to 1.5 Q of the flow rate of permeate water, and the reversible and irreversible resistances were analyzed by applying water backwash five times. Filtration and water backwash were performed for 29.5 and 0.5 min durations, respectively. PO/CF pretreatment was conducted by applying the contact time of 120 s and 25 mg Fe/L doses.

Fig. 7(a) shows the different resistances of membrane fouling resulting from the various pretreatment conditions. Without pretreatment, the resistance after six cycles of filtration increased to 5.85E+12 m^{-1}; with CF, it increased to 2.01E+12 m^{-1}; with PO, it increased to 1.20E+12 m^{-1}; and with PO/CF it increased to 1.05E+12 m^{-1}. Thus, the use of the combined PO/CF treatment yielded the lowest filtration resistance.

Fig. 7(b) shows the reversible and irreversible resistance values determined for the various pretreatment conditions. Physical backwashing was applied five times and the filtration resistances were analyzed after each cycle. The reversible and irreversible resistances gradually increased after each cycle. In the case of filtration without any pretreatment, they increased to 1.73E+12 m^{-1} after five cycles. When the filtration was performed by injecting ozone and FeCl$_3$, the reversible and irreversible resistance values increased to 3.84E+11 m^{-1} and 3.30E+11 m^{-1}, respectively. These values were lower than those in the case without any pretreatment. In the case of combined PO/CF, the reversible and irreversible resistance values were the lowest at 1.66E+10 m^{-1} and 1.09E+11 m^{-1}, respectively.

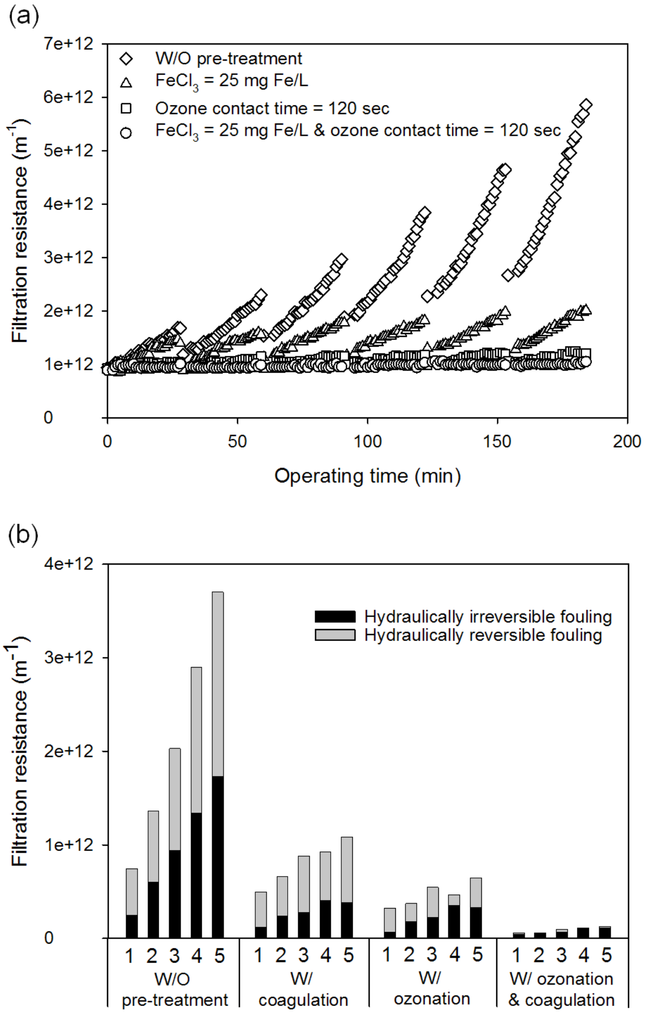

Fig. 7. Variation of: (a) filtration resistance profiles and (b) resistance of hydraulic reversible and/or irreversible membrane fouling with and without ozonation and coagulation/flocculation.

4. Conclusions

To reduce filter fouling during UF for wastewater reuse, we evaluated the treated water quality characteristics, FDR, and reversible and irreversible filtration resistances resulting from the use of ozone and coagulant pretreatments. Ozone pretreatment reduced the levels of turbidity, color, UVA$_{254}$, COD, T-N, and T-P in secondary effluent samples. FTIR showed that polysaccharide proteins and humic substances known to be foulants existed in the secondary effluent. Ozone pretreatment led to increases and decreases in the FTIR bands corresponding to various organic functional groups.

FEEM analysis showed that after PO pretreatment, most of the organic matter in the secondary effluent was oxidized owing to the high reactivity of the ozone. FDR was reduced when PO pretreatment was applied, and the removal efficiency became higher when contact time of 90 and 120 s were used.

On the other hand, the flux results were worse relative to the control case when contact time of 30 and 60 s were used. Thus, the use of an optimal ozone dose is quite important. Also, the combined use of PO and CF pretreatments yielded better performance in terms of FSV. We analyzed the changes in the filtration resistance of UF membranes with and without the use of PO, CF, and combined PO/CF pretreatments. The reduction efficiencies for reversible and irreversible foulants were greatest for the combined PO and

CF pretreatment. Therefore, this combined pretreatment is expected to promote stable membrane operation, because the injection of optimal ozone and coagulant doses contributes to reduce fouling and improved flux.

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