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The performance of contact flocculation–filtration as pretreatment of seawater reverse osmosis

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ABSTRACT

Deep bed filtration has traditionally been used as a pretreatment in seawater desalination. The performance of contact flocculation–filtration (CFF) as pretreatment of seawater reverse osmosis (SWRO) was evaluated in terms of pressure drop through the filter and removal of organics and turbidity. The average turbidity, total suspended solids, and dissolved organic carbon (DOC) of raw seawater were 0.92 NTU, 3.6, and 1.12 mg/L, respectively. The performances of CFF were experimentally evaluated with different flocculant doses (0.5–3.0 mg Fe^{3+}/L) and rapid mixing times (1.7–14.4 s). Here rapid mixing was performed in a spiral flocculation unit which consisted of a PVC tube of length 0.5 m and internal diameters of 0.16 and 0.40 cm. The experimental results show that the filtration rate of 10.0 m/h led to an extensive increase in both head loss (pressure drop) and turbidity as compared to those at filtration rates of 5.0 and 7.5 m/h. The head loss also significantly decreased when the flocculant dose was reduced from 3 to 0.5 mg Fe^{3+}/L. However, the organic matter (26% of DOC) removal was lower at a lower dose of ferric chloride (1.0 mg/L as Fe^{3+}). The removal efficiency of DOC at low concentration of ferric was improved considerably through the improvement of rapid mixing. The application of CFF process also led to a significant decrease in ultrafilter-modified fouling index (UF-MFI).

Keywords: Contact flocculation–filtration; Organic matter; Pretreatment; Seawater

1. Introduction

Membrane fouling is a major problem for efficient operation of seawater reverse osmosis (SWRO). It leads to the degradation of both quantity and quality of produced water, and consequently results in higher maintenance costs [1]. Foulants can be classified into four categories as follows: soluble inorganic compounds, particulates, organics, and microbial products. Fouling by particulates on the membrane results in extra resistance to filtration. Organic fouling is governed by the interactions between the organic foulants themselves and results in the formation of biofilm through activities of micro-organisms on the membrane surface [2–4].

Deep bed filtration has been used as a common pretreatment method in water treatment as well as seawater desalination for suspended solids or particulates removal [5,6]. This filtration process is normally used for the clarification of dilute suspensions of less than 500 mg/L. Particles mainly adhere to the surfaces and introduce the filtration layer

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themselves. With continuing filtration, deposits accumulate within the filter pores and lead to the change of pore geometry and hydrodynamic conditions. Removal of deposits can take place in whole depth of the filter [7]. However, conventional deep bed filtration cannot remove dissolved organic matter which is mainly responsible for reverse osmosis (RO) fouling.

The application of contact flocculation–filtration (CFF) based on deep bed filtration can be a promising pretreatment solution because of its simplicity and relatively lower operation and maintenance cost. In CFF, flocculation of particles and the separation of flocs and particles occur simultaneously within the filter bed itself. Flocculation takes place by the contact of raw water with the flocculant followed by the separation of particles and flocs by the filter medium. To date, CFF studies have been focused on finding the particle removal mechanism and its behaviors in the filter bed [8,9]. However, not much information is available on the optimization of CFF, especially in terms of organic removal.

This paper presents the experimental results of effect of operational conditions (filtration velocity, contact time of flocculant and flocculant dose) on CFF performance with seawater from Chowder Bay, NSW, Australia. Here, the performance of CFF was evaluated in terms of organic removal efficiency and ultrafilter-modified fouling index (UF-MFI) reduction. The detailed organic fractionations were also made in this study. These results of CFF performance were compared with that of in-line flocculation microfiltration (MF) system.

2. Materials and methods

2.1. Materials

2.1.1. Seawater

In this study, seawater was collected from Chowder Bay, Sydney, Australia. The average turbidity, pH, and dissolved organic carbon (DOC) values of seawater used in experiments were 0.92 NTU, 7.8, and 1.12 mg/L, respectively. Average UF-MFI value was 12,795 s/L^2 and total suspended solid (TSS) was 3.6 mg/L.

2.1.2. Chemicals

Ferric chloride ($FeCl_3$), as a flocculant, has high effectiveness in DOC removal. A stock solution (Fe^{3+} = 1,000 mg/L) was prepared and was injected into the CFF by a dosing pump (Cole Parmer Masterflex Pump).

2.1.3. Filter medium

The deep bed filter was packed with sand (as medium). Sand provided from Riversands P/L, Australia was used as the medium in this study and its properties are given in Table 1.

2.1.4. Membranes

The results obtained from submerged MF coupled with in-line flocculation system at a filtration rate of 20 L/m^2 h (correspond to 33.3 mL/min) was used to compare with that of CFF. MF membrane (Cleanfil®-S, PVDF of 0.1 μm, Kolon membrane) used in this study was a hollow fiber module with an effective membrane area (0.10 m^2). The U-type membrane length was 48.5 cm with an outer diameter of 2.0 mm. This membrane was vertically submerged directly into a 6 L reactor.

2.2. Experimental methods

2.2.1. Contact flocculation–filtration

Short-term filtration experiments were carried out with in-line flocculant addition in the filtration column packed with sand. The experimental run was kept short at 6 h. The experimental set-up is shown in Fig. 1(a). The internal diameter of the filtration column was 2.0 cm. It was packed with sand to a depth of 60 cm from the bottom. The filtration velocities and flocculant doses applied were varied from 5.0 to 10.0 m/h and 0.5 to 3.0 mg/L, respectively. The rapid mixing was performed in a spiral flocculator unit which contained a PVC tube. The length of the spiral tube used as rapid mixing zone was kept at 50 cm but the tube diameter was changed from 0.40 to 0.16 cm to change the velocity gradient and retention time of rapid mixing. Mixing times were calculated based on length, diameter of a tube, and flow rate of feed water. Flocculant was added using a dosing pump to rapid mixing unit for contact with feed water. The solution (destabilized water) was then sent through the packed filter column by gravity. To maintain a constant filtration rate in the system, an effluent pump

Table 1
Physical properties of sand

Parameter	Sand
Effective size (mm)	0.55–0.65
Uniformity coefficient	<1.5
Acid solubility	<2%
Specific gravity	2.65
Bulk density (kg/m^3)	1,500

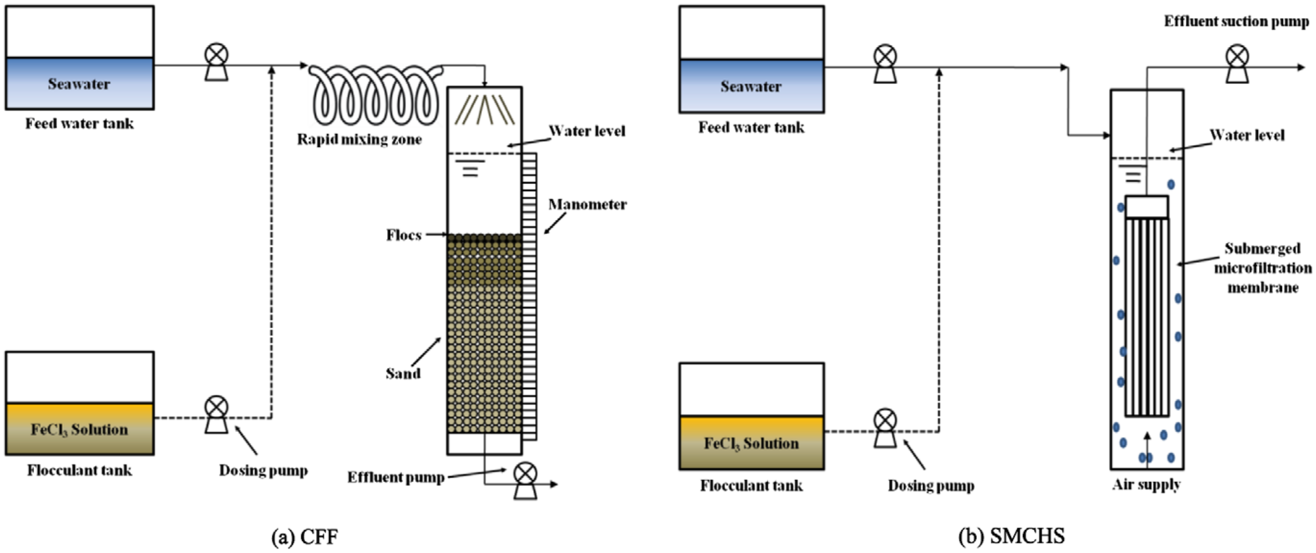

Fig. 1. Schematic diagram of CFF and SMCHS.

was used in the outlet. The filtered samples (filtrates) were collected at the bottom of the column for further analysis.

A few submerged microfiltration coagulation hybrid system (SMCHS) experiments were also conducted to compare its performance with that of CFF. An MF membrane of 0.1 μm pore size was submerged in a 6 L reactor (Fig. 1(b)).

An effective mixing of flocculant with water was achieved by rapid mixing. This is vital for an effective flocculator. An appropriate range of velocity gradient is necessary for proper flocculation. If the G value is too high, the flocs may be sheared but if it is too low, sedimentation may occur within the flocculator [11]. G value in the rapid mixing unit was determined by measuring the head loss across the given length of the spiral tube. The relationship between the head loss and the G value is expressed by Eq. (1).

$$G = \sqrt{\left(\frac{g}{v}\right)\left(\frac{Q}{V}\right)\Delta H} \quad (1)$$

where, G = velocity gradient (s^{-1}), Q = flow rate ($cm^3\,s^{-1}$), V = volume of the flocculator (cm^3), H = head loss through the flocculator (cm H_2O), g = gravitational acceleration ($cm\,s^{-2}$), v = linear flow rate ($cm\,s^{-1}$), and d = internal diameter of the tube (cm). In a tube-type rapid mixing unit, G value was varied by the flow rate and calculated by the empirical relationship (Eq. (2)) established by previous researches [12].

$$G = 6.02\left(\frac{v}{d}\right)^{1.15} \quad (2)$$

The operation parameters used in this study are given in Table 2.

Table 2
Operating condition used in CFF experiments

Filtration rate		Rapid mixing tube length	Tube diameter	Mixing time	Velocity gradient (G)	Flocculant dose
(m/h)	(mL/min)	(cm)	(cm)	(s)	(s^{-1})	(mg Fe^{3+}/L)
5.0	26.2	50	0.40	14.4	72	3.0
						1.0
7.5	39.3		0.40	10.8	101	3.0
						1.0
			0.16	1.7	2,418	1.0
						0.5
10.0	52.4		0.40	7.2	160	3.0
						1.0

The measurement of head loss was observed directly using manometer. The turbidity of raw seawater and filtered seawater was measured by turbidity meter (HANNA, HI 98703) immediately after sampling.

2.2.2. UF-MFI and organic removal

The MFI was measured using ultrafiltration (UF) membranes. The details of UF-MFI measurement are described elsewhere [9]. A fouling index was used to predict how rapidly given feed water will make a fouling on the RO membrane due to colloidal fouling. From this information, an appropriate pretreatment scheme can be suggested. The calculation of UF-MFI value was measured with the filtrate of CFF collected during the first 2 h and the last 4 h. This is to distinguish the filtration stage over the time.

DOC of CFF effluent was measured after filtering the samples through a 0.45 μm filter. The detailed organic fractions were measured by DOC-LABOR Liquid Chromatography–Organic Carbon Detector (LC–OCD). LC–OCD was measured to identify the different classes (polysaccharide, humic substances, building blocks, and low molecular weight neutrals) of organic compounds present in seawater that cause organic fouling. It gives both the qualitative and quantitative detailed information of the organic matter present in seawater before and after treatment.

3. Results and discussion

3.1. Effect of filtration rate

Our previous study with Chowder Bay seawater showed that ferric concentration of 3.0 mg/L was the optimum dose for removing organic matter [10] and this dose was selected for evaluating the effect of filtration rate (5.0, 7.5, and 10.0 m/h) on CFF performance. In this study, there were no differences of head losses in the first 2-h operation among different filtration rates. The increase of filtration rate only resulted in higher head loss after 2 h of operation (Fig. 2(a)). This phenomenon can be explained by the increase of solid loading rate at a higher flow rate and after 2 h of operation, the accumulation of particles in the pores of the filter at different flow rates was high enough to increase the head-loss rate. In addition, the increase of filtration rate also led to particles to infiltrate deeper into the filter bed. Some small particles may have been escaped from the filter. As a result, the turbidity in the effluent at a filtration rate of

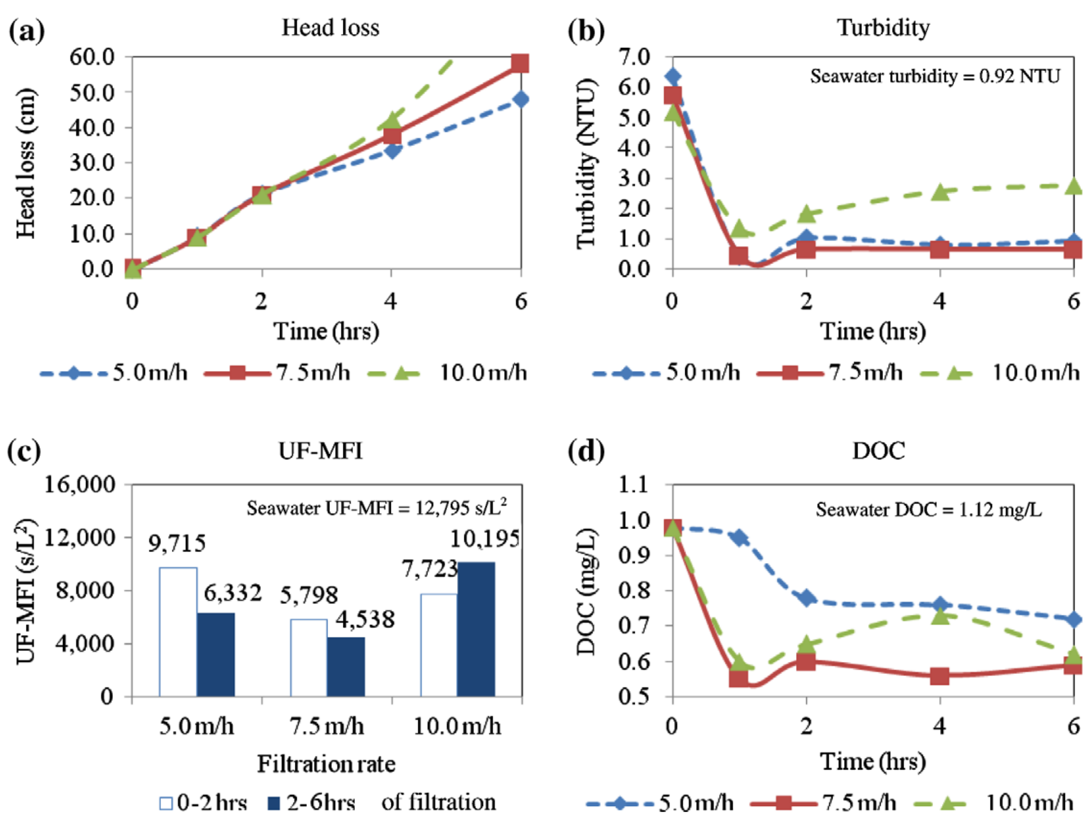

Fig. 2. Effect of filtration velocity on the performance of CFF (3.0 mg of Fe^{3+}/L).

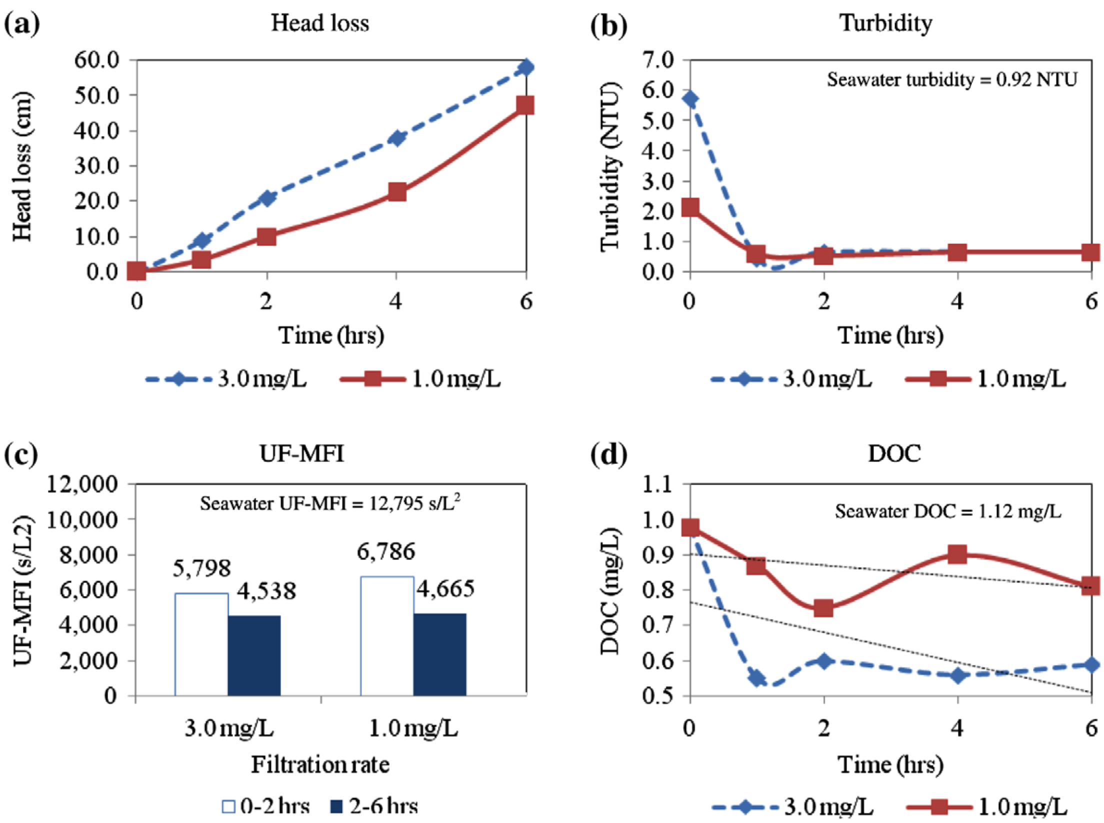

Fig. 3. Effect of flocculant dose (filtration velocity: 7.5 m/h; velocity gradient of rapid mixer G: 101 s^{-1}; contact time in the rapid mixing unit: 10.8 s).

10.0 m/h was remarkably higher than that at filtration rates of 7.5 m/h and 5.0 m/h. The flocculation efficiency in CFF is affected by rapid mixing provided with different velocity gradients (G) and mixing time values. Here the rapid mixing times and G values were varied from 14.4 to 7.2 s and 72 to 160 s^{-1} when the filtration rates increased from 5.0 to 10.0 m/h, respectively (Table 2). As can be seen from Fig. 2(b), turbidities of flocculated seawater after rapid mixing through spiral tube decreased from 6.4 to 5.2 NTU.

The results of experiments show that at lower filtration rates of 5.0 and 7.5 m/h, the UF-MFI reduction improved with time but at a high filtration rate of 10 m/h, the UF-MFI value increased after 2 h of operation (Fig. 2(c)). The increase in UF-MFI could be the result of the increase of turbidity in the effluent. Filtration rate also affected the DOC removal efficiency. The highest DOC removal efficiency was achieved at a filtration rate of 7.5 m/h (Fig. 2(d)).

Table 3
Effect of velocity gradient of rapid mixing device on CFF performance (filtration rate: 7.5 m/h; flocculant dose: 1.0 mg of Fe^{3+}/L)

G value (s^{-1})	101			2,418		
Filtration time (h)	Head loss (cm)	Filtrate turbidity (NTU)	Filtrate DOC (mg/L)	Head loss (cm)	Filtrate turbidity (NTU)	Filtrate DOC (mg/L)
0	0	2.11	1.12	0	3.45	1.12
1	3.5	0.61	0.87	4	0.59	0.62
2	10	0.54	0.75	8.5	0.43	0.75
5	22.5	0.65	0.90	18.5	0.35	0.60
6	47	0.66	0.81	32	0.37	0.76

From these results, filtration rate of 7.5 m/h was found to be the suitable velocity in terms of the removal of turbidity, DOC, and UF-MFI. Therefore, this filtration rate was used in the subsequent experiments.

3.2. Effect of flocculant dose

Fig. 3 shows the effect of flocculant dose at a filtration rate of 7.5 m/h and rapid mixing with a velocity gradient (G) value of 101 s^{-1}. In this experiment, the flocculant dose was decreased from 3.0 to 1.0 mg of Fe^{3+}/L. The lower concentration of ferric led to lower head loss. After a contact with 1.0 mg/L of flocculant, through a spiral rapid mixing unit, the turbidity was lower than that after a contact with 3.0 mg/L of flocculant (Fig. 3(b)). The results also show that the turbidity after filtration was constant (approximately 0.65 NTU) with both concentrations of flocculant. There was also not much of difference in UF-MFI value when a low concentration of ferric was used. However, the DOC removal was very low (<27%) when a low concentration of ferric was applied.

3.3. Effect of velocity gradient

G value of rapid mixing unit is affected by tube diameter. It is inversely proportional to tube diameter. The linear velocity through tube is increased in smaller diameter tubes at a given filtration rate. In this study, the internal tube diameter (d) of the tube used as a rapid mixing device was decreased from 0.40 to 0.16 cm. As the tube diameter decreased to 0.16 cm, G value increased approximately 24 times from 101 to 2,418 s^{-1}. Due to the increase of velocity gradient, DOC removal efficiency improved significantly; from about 26–44% although the rapid mixing time was as low as 1.7 s. In addition, when G value was increased to 2,418 s^{-1}, the turbidity in the effluent was superior of less than 0.4 NTU. This also led to a lower pressure drop during an operation time of 6 h (Table 3).

3.4. Organic fractionation

Low pressure membrane systems such as MF system have been widely used as a pretreatment to RO as they can remove macromolecules, bacteria, and discrete particles from feed water and can help to reduce RO membrane fouling. CFF is more cost-effective than MF and can be considered as an alternative pretreatment for RO. In this study, the performance of CFF was compared with that of SMCHS in terms of the detailed organic fractions. Our previous jar test with the same seawater showed that there was no effective flocculation at the low concentration of 0.5 mg of Fe^{3+}/L [10]. At this concentration, flocs size was less than 2 μm and flocs could not be able to be observed by naked eyes. In the experiment with CFF, the flocculation performance was improved by the incorporation of the rapid mixing through the use of spiral flocculator. The CFF performance was compared with the result we obtained previously with SMCHS [10]. The total DOC removal efficiency by CFF, in comparison with SMCHS, was nearly the same. Although the hydrophobic compound removal by CFF was lower, CFF could remove a significant portion of hydrophilic compounds (Table 4). In particular, the removal efficiency of humic substances by CFF reached up to 74.5%. This analysis needs to be investigated further to obtain a concrete conclusion.

Table 4
Detailed organic fractions by CFF at optimum condition compared with SMCHS

	DOC			Hydrophilic DOC			
Sample	Total	Hydrophobic	Hydrophilic	Bio-polymers	Humics	Building blocks	Neutrals
Seawater (mg/L)[a]	1.12	0.23	0.89	0.12	0.47	0.22	0.08
Treatment by CFF* (mg/L)[a]	0.47	0.08	0.39	0.11	0.12	0.11	0.05
Removal efficiency by CFF (%)[b]	58.0	65.2	56.2	8.3	74.5	50.0	37.5
Seawater (mg/L)[a]	1.29	0.46	0.83	0.13	0.43	0.18	0.09
Treatment by SMCHS** (mg/L)[a]	0.57	0.02	0.55	0.05	0.25	0.14	0.04
Removal efficiency by SMCHS (%)[b]	55.8	95.7	33.7	61.5	41.9	22.2	55.6

[a]Concentrations of the different organic fractions in seawater.
[b]Removal efficiencies of different organic fractions in seawater after treatment.
*CFF (contact flocculation filtration) at 7.5 m/h (39.3 mL/min), $G = 2418\ s^{-1}$, Fe^{3+}: 0.5 mg/L.
**SMCHS (submerged membrane coagulation hybrid system) at 20 L/m^2 h (33.3 mL/min), Fe^{3+}: 0.5 mg/L.

4. Conclusion

The performance of CFF was evaluated at different operation conditions. As filtration rate increased from 5.0 to 10.0 m/h, the pressure drop (head loss) increased and removals in terms of turbidity and DOC declined. The incorporation of rapid mixing device prior to CFF had a positive effect on the improvement of filtration quality. At a low velocity gradient ($101\,s^{-1}$) of the rapid mixing unit, the reduction of flocculation dose led to inferior DOC removal efficiency. However, at the same operation conditions, the increase of the velocity gradient (to $2{,}418\,s^{-1}$) enhanced the in-line flocculation performance, and in particular, the turbidity and DOC removal efficiencies were improved. At low concentration of 0.5 mg of Fe^{3+}/L, CFF could remove both hydrophobic and hydrophilic compounds. The organic removal of CFF was comparable to that with SMCHS.

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