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Microcoagulation improved the performance of the UF–RO system treating the effluent from a coastal municipal wastewater treatment plant: a pilot-scale study

Tong Yu, Chenlu Xu, Feng Chen, Haoshuai Yin, Hao Sun, Lihua Cheng Mand Xuejun Bi

ABSTRACT

Microcoagulation has recently been considered as a promising pretreatment for an ultrafiltration (UF) process from numerous studies. To investigate the effects of microcoagulation on the performance of the UF–reverse osmosis (RO) system treating wastewater with high and fluctuant salinity, different dosages of coagulant (poly-aluminum chloride) were added prior to the UF unit in a pilot-scale UF–RO system for a 10-week period operation. Microcoagulation obviously improved the contaminant removal and cleaning efficiencies, including water backwash, chemical enhanced backwash and cleaning in place processes. Organic fouling was dominated during the initial stage of the RO membrane fouling. The microbial communities of water samples and foulant on the RO membrane were similar to those of seawater and foulant on the RO membranes from seawater RO plants. The microbial community of the foulant on the membrane was similar to that of UF permeate and RO concentrate. These results demonstrated that microcoagulation could improve the performance of the UF–RO system treating the effluent with high and fluctuant salinity from a coastal municipal wastewater treatment plant.

Key words | cmWWTP, membrane cleaning, membrane fouling, microcoagulation, pilot UF-RO system

HIGHLIGHTS

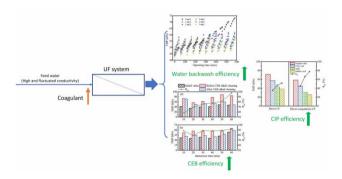
- A pilot-scale ultrafiltration (UF)-reverse osmosis (RO) system with microcoagulation was applied.
- Microcoagulation-UF had a better efficiency of contaminant removal than direct UF.
- Microcoagulation improved the efficiency of water backwash, chemical enhanced backwash and cleaning in place.
- Organic fouling was dominated in the initial stage of the RO membrane fouling.
- The microbial community of the foulant was similar to that of UF permeate and RO concentrate.

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GRAPHICAL ABSTRACT



INTRODUCTION

Wastewater reclamation is one of the important means to overcome the water crisis. In recent years, to obtain highquality reclaimed water, an ultrafiltration (UF)-reverse osmosis (RO) process has been developed as an effective approach due to its stable permeate quality and automatic operation (Tang et al. 2016; Anis et al. 2019; Ezugbe & Rathilal 2020). Despite the wide applicability of the UF-RO process, the fouling of UF and RO membranes adversely impacts the performance of the UF-RO system, which severely hinders the development of the UF-RO process (Flemming 1997; Schneider et al. 2005; Gao et al. 2011; Xu et al. 2019).

The feed water (FW) composition is one of the major factors governing the system performance and membrane fouling. Therefore, the pretreatment of the UF-RO process should also consider the difference in FW quality (Khan et al. 2013b, 2014). In a coastal municipal wastewater treatment plant (cmWWTP), the salinity of the effluent is usually higher than that of other plants and fluctuates periodically due to seawater intrusion. Therefore, when using the UF-RO process to treat the effluent from cmWWTP, additional considerations should be taken to the performance and pretreatment of the UF-RO system.

To optimize the performance of the UF-RO system and alleviate membrane fouling, several pretreatment methods were investigated and applied. Coagulation, which could remove colloid and high-molecular-weight organics, was recognized as an effective pretreatment method for alleviating the membrane fouling, and continuous attention was paid by recent studies (Humbert et al. 2007; Huang et al. 2017; Li et al. 2019). Comparing with the traditional coagulation process, microcoagulation has advantages of less coagulant dosage, shorter coagulation time and less footprints (Guigui et al. 2002; Yu et al. 2015; Ferrer et al. 2016; Aly et al. 2018; Xu et al. 2020). It seems that the microcoagulation is more suitable for the pretreatment of UF. However, in the UF-RO system treating cmWWTP effluent with high and fluctuant salinity, there were few reports on the effect of microcoagulation on pollutant removal and membrane fouling alleviation, especially in pilot-scale studies.

In this study, a pilot-scale UF-RO system was applied to investigate the influences of microcoagulation-UF (mcUF) on contaminant removal in the cmWWTP effluent with high and fluctuant salinity. Furthermore, the backwash and chemical cleaning efficiencies of UF and mcUF were compared. After 10 weeks of operation of the UF-RO system, the UF and RO membranes were autopsied to evaluate initial-stage membrane foulants (e.g. inorganics, organics and bacteria) under this water quality condition.

METHODS

Description of cmWWTP and the pilot-scale UF-RO system

A pilot-scale UF-RO system was conducted at a cmWWTP, which was located in Qingdao, Shandong Province, China. The cmWWTP adopted anaerobic-anoxic-oxic moving-bed biofilm reactor as a secondary treatment process for the biological removal of nitrogen and phosphorus (Supplementary Material, Figure S1). The FW for the pilotscale UF-RO system was collected from cmWWTP effluent with additional coagulation, filtration and UV disinfection treatments. The pilot-scale UF-RO system, as shown in Figure 1, has a capacity of 48 m³/day and a water recovery rate of 70%. The characteristics of the UF (inge dizzer[®]) 5000 plus 0.9MB50) and RO membranes (DOW FILMTEC™ LC HR-4040) used in the pilot system are shown in Supplementary Material, Table S1. The RO array is two stages with two pressure vessels in parallel followed by one single pressure vessel. One pressure vessel contains six 4-inch spiral-wound modules housed in series (Supplementary Material, Figure S2).

Experimental setup

The experimental setup of microcoagulation, water backwash, chemical enhanced backwash (CEB) and cleaning in place (CIP) processes is shown in Figure 1. Polyaluminum chloride (PAC) was used as a coagulant for microcoagulation. During the experimental period, different doses of PAC (1, 2, 3, 4 and 5 mg/L, respectively) were added prior to the UF unit, with a retention time of 60 s. The water backwash was performed every 45 min with a flux of 230 L/(m²·h) and a duration of 40 s. The CEB process was performed on a daily basis. The CEB process sequence was as follows: alkaline cleaning (NaOH, pH 12, 40 s), alkaline immersion (10-60 min), water backwash (40 s), acid cleaning (H₂SO₄, pH 2, 40 s), acid immersion (40 min) and water backwash (40 s). UF permeate (UFP) was used as cleaning water for the water backwash and CEB process. RO permeate was used as cleaning water for the CIP process. The CIP process sequence was as follows: acid treatment (citric acid, pH 2, cycle 3 h - immersion 4 h), followed by water treatment (rinse to neutral), then alkaline treatment (NaOH, pH 12, cycle 3 h - immersion 14 h), and a second water treatment (rinse to neutral), then a second acid treatment (H₂SO₄, pH 2, cycle 3 h - immersion 12 h) and finally a third water treatment (rinse to neutral).

Sample preparation

Water samples

Three different water samples, including FW, UFP and RO permeate and RO concentrate (ROC), were collected every 1 or 2 days from the pilot-scale UF-RO system. Water samples were filtered using a 0.2-µm nylon membrane (Whatman, UK) to eliminate suspended solids and stored at 4 °C prior to analyses.

Membrane samples

A laboratory-scale UF membrane filament, which was in parallel with the pilot UF module, was collected (Supplementary Material, Figure S3). Three different kinds of solutions (1 M HCl, 1 M NaOH and ultrapure water (UPW)) were used to dissolve the deposits on the fouled UF membranes. Three 20-cm UF membrane filaments were, respectively, soaked in 300 mL each of 1 M HCl, 1 M NaOH and UPW for 2 h with moderate stirring. The pH of the three soaking solutions (with desorbed UF membrane foulants) was adjusted to 7.0 using NaOH and HCl and then was filtered immediately by 0.2-µm nylon membranes (Whatman, UK). The three obtained permeate samples were referred to as

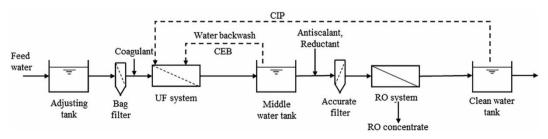


Figure 1 | Schematic diagram of the major process in the pilot UF-RO system.

 $S_{\rm HCl}$, $S_{\rm NaOH}$ and $S_{\rm UPW}$ (Tang et al. 2014). Six RO membrane modules in the first stage were collected, drained and weighed. The first RO membrane module in the first stage was collected and stored at 4 °C to minimize the changes of constituents on the RO membrane surface (Supplementary Material, Figure S2). The foulants at the inlet and outlet parts of the fouled RO membrane were collected by physical scrapping. The inlet part of the RO membrane (M1) was nearest to RO influent and the outlet part of the RO membrane (M2) were nearest to ROC.

Analytical methods

Basic index

Conductivity (Cond.), pH, transmembrane pressure (TMP) and membrane flux were measured by an online monitor of the UF-RO system. Salt rejection was defined by Equation (1). The recovery of membrane specific flux (R_{SF}) was defined by Equation (2).

$$\begin{aligned} Salt\,rejection &= (Cond._{influent} - Cond._{permeate})/Cond._{influent} \\ &\times 100\% \end{aligned}$$

(1)

$$R_{\rm SF} = \rm SF/SF_0 \times 100\% \tag{2}$$

where SF is the specific flux after cleaning, L/(m² h kPa), and SF_0 is the initial specific flux, $L/(m^2 h kPa)$.

Turbidity was measured by an SGZ-20B portable turbidimeter (Mingbolm, China). Dissolved organic carbon (DOC) and total nitrogen (TN) were measured by a multi N/C 2100 analyzer (Analytik Jena, Germany). UV absorbance at 254 nm (UV $_{254}$) was measured by a Uvmini-1240 spectrophotometer (Shimadzu, Japan).

Morphological analysis

Photographs of the UF and RO membranes were taken by a camera. The surface morphology and interface element analysis of fouled RO membranes were conducted by quanta 250 FEG scanning electron microscopy (SEM; FEI, USA) equipped with an energy dispersive spectroscopy (EDS).

Loss on ignition

Loss on ignition (LOI) analysis was conducted for the fouled RO membrane. The foulants on the membranes were dried at 110 °C and heated to 550 °C for 12 h. The mass loss and the remaining mass on ignition at 550 °C represented the content of organics and inorganics, respectively (Tan et al. 2017).

Elemental analysis

An inductively coupled plasma optical emission spectroscopy (ICP-OES; Varian VISTA-MPX, USA), and a 761 Compact Ion Chromatography (IC, Metrohm, Switzerland) were used for inorganic element analysis. The element deposition ratio (EDR) as described by Tang et al. (2014)is defined as the percentage of the amount of an inorganic element deposited on the membrane over the total amount of the inorganic element in the RO influent for 70-day operation (Equation (3)).

$$EDR = (C_{im} \times P_i) / (C_{iw} \times Flux \times 24 \text{ h} \times 70 \text{day}) \times 100\%$$
 (3)

where $C_{\rm im}$ is the content of inorganic foulants on the membrane measured by LOI analysis, mg/m^2 ; P_i is the proportion of an inorganic element in the total inorganic matter measured by SEM-EDS, %; Ciw is the concentration of an inorganic element in the RO influent measured by ICP-OES, mg/L; and Flux, L/(m² h).

Microbial analyses

Foulants from 5 × 5 cm² membrane samples were suspended in 25 mL of phosphate-buffered saline. Heterotrophic plate counts (HPCs) of membrane foulant samples and water samples were performed on R2A medium (Sigma-Aldrich, USA) according to Reasoner & Geldreich (1985).

The genomic DNA was extracted from the samples of 500 mL of FW, UFP, ROC and 50 mg of foulants on RO membranes using the OMEGA D5525-01 E.Z.N.A.™ Water DNA kit (USA) according to the manufacturer's protocols. The V4 region of 16S rRNA gene was PCR amplified using the primer pairs (515F/806R) (Caporaso et al. 2012). Pyrosequencing of 16S rRNA genes was performed on the Illumina MiSeq platform (USA). Sequences were processed by Mothur v.1.32.0 in order to reduce the noise and sequencing artifacts, as described by Pang et al. (2016). Sequences were clustered into operational taxonomic units based on 97% sequence similarity by comparison to the Silva reference database (db119) (July 2014).

RESULTS AND DISCUSSION

Water characteristics

Water characteristics through the pilot UF-RO system, in terms of pH, conductivity, DOC, UV₂₅₄, TN, turbidity, metal ions and anions, were monitored in order to investigate the relationship between the FW characteristics and foulants on membranes (Supplementary Material, Table S2). The conductivity, sodium, chloride and sulfate in the FW of the UF-RO system (cmWWTP effluent) were $1,811-5,420 \,\mu\text{s/cm}$, $414.1-856.5 \,\text{mg/L}$, 1,579.8 mg/L and 198.8-349.2 mg/L, respectively, which were significantly higher than those of other common mWWTPs (Xu et al. 2010; Chon et al. 2012; Ayache et al. 2013; Wang et al. 2019; Tong et al. 2020), which could be resulted by the periodical seawater intrusion of this cmWWTP. The UF membrane removed turbidity to a certain extent but had no ability to remove organic matter and ions. The RO membrane removed all kinds of pollutants effectively, indicating that the UF-RO system could

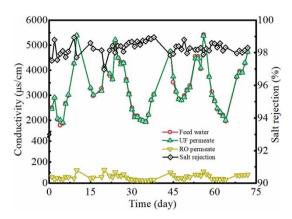


Figure 2 | The conductivity and salt rejection of the UF-RO system during the operation period with the salinity fluctuation of water quality.

operate normally under this water quality condition (Supplementary Material, Table S2).

Effect of salinity fluctuation on the UF-RO system

With FW conductivity fluctuating periodically, the RO permeate conductivity and salt rejection fluctuated slightly (Figure 2). The data showed that the flux decreased approximately from 14 to 10 L/(m²·h) and the salt rejection decreased approximately from 99 to 98%, as the conductivity of FW increased from 1,811 to 5,420 us/cm, respectively (Figure 3). The results indicated that the UF-RO system could guarantee the permeate quality (salt rejection above 98%) under such a conductivity fluctuation condition. Therefore, the wastewater reclamation UF-RO

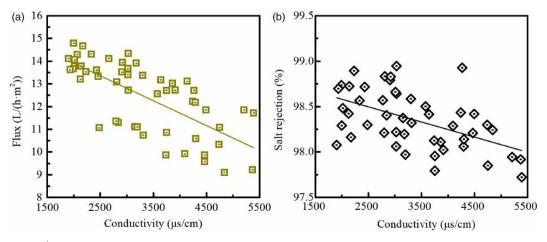


Figure 3 | Effect of FW conductivity on flux (a) and salt rejection (b).

process could be applied for the production of high-quality reclaimed water. However, if the conductivity of the FW increases further, it may affect the performance stability of the UF-RO system and the water quality of the RO permeate (Figure 3).

Effect of microcoagulation on the removal of contaminants

Coagulation is a widely used conventional technique in reclaimed water treatment processes for removing colloid and high-molecular-weight organics (Zhao et al. 2014; Yu et al. 2016). Comparing with direct UF, except for TN, mcUF had better removal efficiencies of turbidity, DOC and UV₂₅₄, which was more favorable for the subsequent RO operation (Figure 4). These results were consistent with another study on mcUF for treating activated carbon filter backwash water (Zhang et al. 2017). The removal rate of turbidity by mcUF with all the given PAC dosages was consistently higher than that of direct UF (66.1%), where at the dosage of 4 mg/L PAC, the removal rate of turbidity by mcUF reached the highest of 91.7%. Similarly, the removal rates of DOC and UV₂₅₄ by direct UF were 6.2 and 4.8% respectively, while the removal rates of DOC and UV₂₅₄ by mcUF with different PAC dosages were consistently higher.

It should be noted that the conductivity had an effect on the removal efficiency of organic matter by coagulation. In this study, the effect of conductivity on the removal of organic matters by microcoagulation using the actual cmWWTP effluent with two kinds of conductivities (4,840 and 2,220 µs/cm) was conducted via the laboratory-scale experiment. It was found that higher conductivity could enhance the removal efficiency of organic matter by coagulation in certain degree (data not shown). In the process of pilot-scale test, the conductivities corresponding to different coagulant concentrations are shown in Supplementary Material, Table S3. It could be noted that with the increase

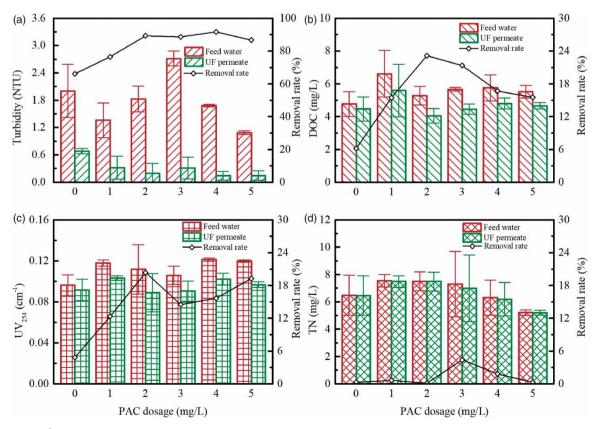


Figure 4 Removal of turbidity (a), DOC (b), UV₂₅₄ (c), and TN (d) by direct UF (0 mg/L) and mcUF with different dosages of PAC (1–5 mg/L).

of coagulant concentration after 2 mg/L, the removal efficiency of organic matter decreased, which may be caused by the decrease of conductivity.

Effect of microcoagulation on UF cleaning

To investigate the effect of microcoagulation on UF performance and the efficiency of cleaning, mcUF with different dosages of PAC was run for 10 cycles (filtration for 45 min and backwash with UFP for 40 s), respectively. As shown in Figure 5, the TMP of direct UF remarkably increased after each cycle, while the TMP of mcUF with different dosages of PAC showed a slighter increase. After 10 cycles, the TMP of direct UF was significantly higher than that of mcUF.

The increase of TMP for mcUF with different PAC dosages was higher than direct UF within one cycle (Figure 5). However, the water backwash efficiency of mcUF was significantly better than that of direct UF. Conductivity had effect on the turbidity and organic matters removal by microcoagulation. The water backwash of UF membrane was also affected by conductivity fluctuation. Therefore, there was no obvious rule for the efficiency of water backwash with different coagulant concentrations. However, it was clear that microcoagulation could improve the efficiency of water backwash. Taking dosage of 4 mg/L PAC as an instance, although the TMP of mcUF increased approximately 10 kPa within one cycle, which was much higher than that of direct UF of approximately 5 kPa, most

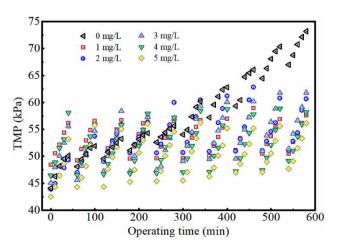


Figure 5 | TMP variation of direct UF and mcUF with different PAC dosages in 10 cycles.

of them could be removed by the water backwash more easily. The R_{SF} maintained above 99% during most cycles, which were higher than those of direct UF of 95% (Supplementary Material, Figure S4). Similarly, researchers reported that the floc cake layer on the membrane surface formed by coagulation could be easily removed by backwashing and flushing (Dong et al. 2007; Xu et al. 2020). This result indicated that the performance of mcUF was more stable than that of direct UF.

The effects of CEB alkali cleaning on mcUF (PAC 4 mg/L) and direct UF with different immersion times were compared. With the alkali immersion time increased, the R_{SF} of direct UF increased from 60% to nearly 100%, which indicated that the foulant on the UF membrane surface could not be easily removed by alkali cleaning with short-time immersion. While, the R_{SF} of mcUF remained stable at approximately 90% even with immersion time of 10 min (Figure 6). These results showed that the CEB alkali cleaning efficiency of mcUF was more stable and had a better cleaning effect with shorter immersion time.

CIP is a more complex cleaning process, which was applied when the TMP could not be recovered by the water backwash and CEB process. The effects of the CIP process on mcUF (PAC 4 mg/L) and direct UF were

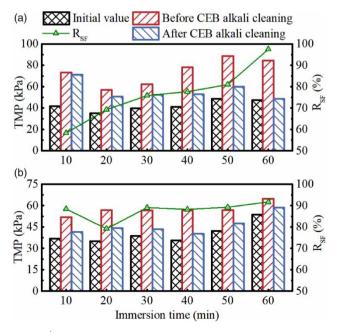


Figure 6 | The TMP decrease and R_{SF} of direct UF (a) and mcUF (b) by CEB alkali cleaning.

compared. The $R_{\rm SF}$ of each step in CIP of mcUF was much better than that of direct UF. The R_{SF} of the whole CIP process of mcUF was nearly 30% higher than that of direct UF (Figure 7). This result showed that microcoagulation could improve the cleaning of CIP effectiveness of UF significantly.

Membrane fouling analyses

UF membrane fouling analysis

Three different kinds of solutions (1 M HCl, 1 M NaOH and UPW) were used to dissolve the deposits on fouled UF membranes. The organic contaminants on the UF membrane obtained by immersion in pure water are significantly lower than those of the S_{HCl} and S_{NaOH} . The absorbance of UV_{254} and the concentration of TN in the S_{HCl} are significantly lower than those of the S_{NaOH} (Table 1). The result of simple immersion experiment also showed that alkali cleaning is more effective than acid cleaning for the organic contaminant removal. In terms of inorganic contaminants,

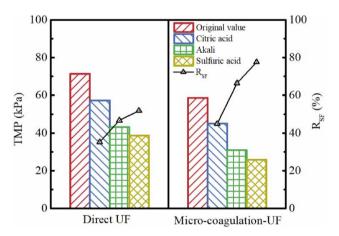


Figure 7 The TMP decrease and R_{SF} of direct UF (a) and mcUF (b) by the CIP process.

Table 1 Organic foulants on the UF membrane

Filtrate type	UV ₂₅₄ (cm ⁻¹)	DOC (mg/L)	TN (mg/L)
$S_{ m UPW}$	0.006	1.68	0.14
$S_{ m NaOH}$	0.113	8.44	1.29
S_{HCl}	0.013	8.81	0.24

acid cleaning could remove Ca²⁺ and Mg²⁺ effectively (Supplementary Material, Table S4).

RO membrane fouling analysis

Microcoagulation could improve the removal efficiency of organic matters. Theoretically, it could improve the performance of the RO system. However, due to the short experimental period, the RO system had been running well, so the data could not reflect the effect of microcoagulation. To share more information on the RO membrane fouling in the initial stage of operation under the water quality with high and fluctuated conductivity, the RO membrane autopsy and analyses were conducted in this study. After 10 weeks of operation of the UF-RO system, to investigate RO fouling in the initial stage, the six RO membrane modules in the first stage were collected, drained and weighed. The first membrane module was the heaviest among all the six membrane modules (data not shown). Therefore, the first membrane module was selected for anatomy to investigate the most severe fouling membrane. Macroscopic observation showed that there were significantly more foulants in the inlet part (M1) of RO membrane than those in the outlet part (M2) (Supplementary Material, Figure S5). Similarly, microscopy observation also showed more foulants at the inlet part (Supplementary Material, Figure S5).

The contents of the organic and inorganic components of the foulants were measured by LOI analysis with data summarized in Table 2. The contents of the organic components of M1 and M2 were 1.55 ± 0.33 and 0.81 ± 0.31 g/m², respectively, accounting for approximately 98.1 ± 0.2 and $96.3 \pm 0.7\%$ of the foulant dry weights, whereas the contents of the inorganic component were 0.03 ± 0.01 and 0.03 ± 0.02 g/m². The LOI analysis showed that the organic content in the inlet part

Table 2 | The content of organic and inorganic components on the RO membrane

	M1	M2
Dry weight (DW, g/m ²)	$1.58\pm0.34^{\rm a}$	0.81 ± 0.33
Organic content (g/m²)	1.55 ± 0.33	0.78 ± 0.31
Organic content (% of DW)	98.1 ± 0.2	96.3 ± 0.7
Inorganic content (g/m²)	0.03 ± 0.01	0.03 ± 0.02
Inorganic content (% of DW)	1.9 ± 0.2	3.7 ± 0.7

^aErrors indicate the standard deviation of three replicates of the deposit tested

(M1) was twice as high as that in the outlet part (M2). In terms of the total amount, organic fouling was the major problem for the RO membrane. These results were accorded with the results of studies on RO membranes in full-scale plants (Tang et al. 2014, 2016).

In terms of the deposition ratio of the elements, consistent with other anatomical analyses, Fe could deposit on the RO membrane much more easily than other elements (Supplementary Material, Table S5) (Tang et al. 2014; Tan et al. 2017). Baker & Dudley (1998) recognized that the presence of Fe bonding with organic matter and extracellular polymeric substances (EPS) was a major contributor for membrane fouling. For this reason, the removal of Fe in the RO influent by pretreatment would be crucial for the performance of the UF-RO system.

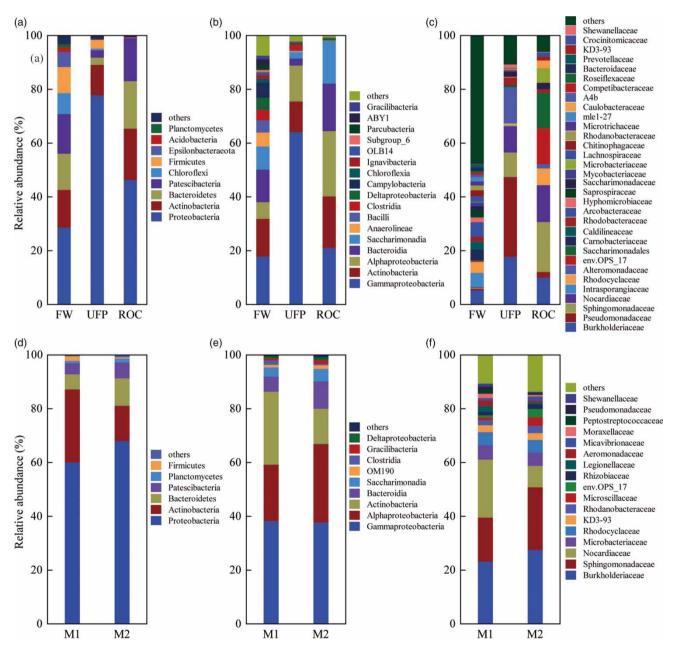


Figure 8 | Microbial communities of the water samples (FW, UFP and ROC) and the membrane samples (M1 and M2) at the phylum level (a and d), class level (b and e) and family level (c and f). 'Others' in the legend indicates the sum of unidentified bacteria and other minor bacteria populations.

HPCs of the water samples and RO membrane samples were performed. The HPC values of FW, UFP and ROC were $1.7 \times 10^2 \pm 7.2 \times 10^1$, $5.2 \times 10^1 \pm 6.0 \times 10^0$ and $2.5 \times 10^0 \times 10^0$ $10^2 \pm 4.5 \times 10^1$ CFU/mL, respectively. Similar to the organic contaminant on the RO membrane, the HPC of M1 (2.9 \times $10^6 \pm 4.6 \times 10^5$ CFU/cm²) is twice as high as that of M2 $(1.3 \times 10^6 \pm 2.9 \times 10^5 \text{ CFU/cm}^2)$ (Supplementary Material, Table S6).

Regarding the structure of microbial communities in the water samples (FW, UFP and ROC) and the RO membrane samples (M1 and M2), triplicate sequencing results are averaged and shown in Figure 8. Proteobacteria was predominant in all water and membrane samples (Figure 8(a) and 8(d)). In terms of the water samples, Gammaproteobacteria, Actinobacteria and Alphaproteobacteria were three dominant classes (Figure 8(b)). The microbial community structure of the water samples was closer to that of seawater, rather than that of the secondary effluent of mWWTP (Khan et al. 2013a, 2013b; Pang et al. 2016). After UF treatment, the microbial diversity of the water decreased significantly (Supplementary Material, Table S7). Meanwhile, the microbial community structure also changed significantly (Figure 8(a)-8(c)). At the family level, Burkholderiaceae, Pseudomonadaceae, Sphingomonadaceae and Nocardiaceae were the four dominant families in the UFP and ROC (Figure 8(c)). Similar to the water samples, Gammaproteobacteria, Actinobacteria and Alphaproteobacteria were the three dominant classes, which are obviously different from the microbial community of the foulant on the RO membrane from wastewater treatment RO plant, but similar to that of seawater RO plant (Figure 8(e)) (Khan et al. 2014). At the family level, similar to the samples of the UFP and ROC, Burkholderiaceae, Pseudomonadaceae, Sphingomonadaceae and Nocardiaceae were the four dominant families on the RO membrane (Figure 8(f)).

The Bray-Curtis dissimilar heatmap showed that the dissimilarities of two membrane parts (M1 and M2) were very low (Figure 9). The microbial communities of UFP and ROC were clustered with that of RO membrane foulant rather than with the FW. However, many researchers reported that the microbial communities significantly differed from the UFP to the RO membranes (Ayache et al. 2013; Khan et al. 2015; Tan et al. 2017). Yu et al. (2017) reported that the microbial community of RO membrane foulant after

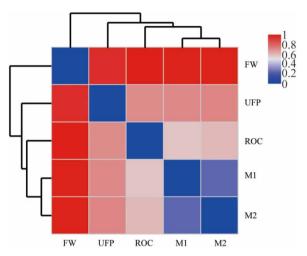


Figure 9 | Bray-Curtis dissimilar heatmap of the water samples and the membrane samples. The Bray-Curtis index ranges from 0 to 1.0, indicating that two samples are composed of the same species. 1 indicates that two samples do not have any of the same species.

membrane chemical cleaning was closer to the water sample than that of the non-cleaned membrane surface. It was speculated that the bacteria deposited on the membrane surface at the initial stage was closer to that of the water sample. This study proved that in the initial stage of membrane fouling, the bacteria on the membrane surface mainly deposited and enriched from the RO FW (UFP). With the increase of operation time, the microbial community on the membrane surface may be gradually differed from that of water.

CONCLUSIONS

In this work, we evaluated the effect of microcoagulation on the performance of the UF-RO system treating the effluent with high and fluctuant salinity from a cmWWTP using a pilot-scale system. mcUF had better removal efficiencies of contaminants than direct UF, which was favorable for the subsequent RO operation. Meanwhile, microcoagulation remarkably improved the cleaning efficiencies of water backwash, CEB and CIP processes. Thus, microcoagulation could improve the performance of the UF-RO system treating the effluent with high and fluctuant salinity from a cmWWTP. The RO membrane anatomical analysis showed that organic fouling was dominated in the initial stage of the RO membrane fouling. The microbial communities of water samples and foulant on the RO membrane were similar to those of seawater and foulant on the RO membranes from seawater RO plants. The microbial community of the foulant on the membrane was similar to that of UFP and ROC.

ACKNOWLEDGEMENTS

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

REFERENCES

- Aly, S. A., Anderson, W. B. & Huck, P. M. 2018 Biological filtration with and without prior in-line coagulation to reduce UF fouling by secondary effluent. Journal of Water Reuse and Desalination 8 (2), 176-191.
- Anis, S. F., Hashaikeh, R. & Hilal, N. 2019 Reverse osmosis pretreatment technologies and future trends: a comprehensive review. Desalination 452, 159-195.
- Ayache, C., Manes, C., Pidou, M., Croue, J.-P. & Gernjak, W. 2013 Microbial community analysis of fouled reverse osmosis membranes used in water recycling. Water Research 47 (10), 3291-3299.
- Baker, J. S. & Dudley, L. Y. 1998 Biofouling in membrane systems - a review. Desalination 118 (1-3), 81-89.
- Caporaso, J. G., Lauber, C. L., Walters, W. A., Berg-Lyons, D., Huntley, J., Fierer, N., Owens, S. M., Betley, J., Fraser, L. & Bauer, M. 2012 Ultra-high-throughput microbial community analysis on the Illumina HiSeq and MiSeq platforms. The ISME Journal 6 (8), 1621-1624.
- Chon, K., Cho, J., Shon, H. K. & Chon, K. 2012 Advanced characterization of organic foulants of ultrafiltration and reverse osmosis from water reclamation. Desalination 301, 59-66.
- Dong, B.-Z., Chen, Y., Gao, N.-Y. & Fan, J.-C. 2007 Effect of coagulation pretreatment on the fouling of ultrafiltration membrane. Journal of Environmental Sciences 19, 278-283.

- Ezugbe, E. O. & Rathilal, S. 2020 Membrane technologies in wastewater treatment: a review. Membrane 10 (5), 89.
- Ferrer, O., Dekker, R., Mesa, J., Martín-Alonso, J., Cortina, J. L. & Gibert, O. 2016 Micro-coagulation effects on direct ultrafiltration of challenging raw river water. Journal of Chemical Technology Biotechnology 91 (12), 2959-2973.
- Flemming, H.-C. 1997 Reverse osmosis membrane biofouling. Experimental Thermal and Fluid Science 14 (4), 382-391.
- Gao, W., Liang, H., Ma, J., Han, M., Chen, Z.-L., Han, Z.-S. & Li, G.-B. 2011 Membrane fouling control in ultrafiltration technology for drinking water production: a review. Desalination 272 (1-3), 1-8.
- Guigui, C., Rouch, J. C., Durand-Bourlier, L., Bonnelye, V. & Aptel, P. 2002 Impact of coagulation conditions on the in-line coagulation/UF process for drinking water production. Desalination 147 (1-3), 95-100.
- Huang, B.-C., Guan, Y.-F., Chen, W. & Yu, H.-Q. 2017 Membrane fouling characteristics and mitigation in a coagulationassisted microfiltration process for municipal wastewater pretreatment. Water Research 123, 216-223.
- Humbert, H., Gallard, H., Jacquemet, V. & Croue, J.-P. 2007 Combination of coagulation and ion exchange for the reduction of UF fouling properties of a high DOC content surface water. Water Research 41 (17), 3803-3811.
- Khan, M. T., de O. Manes, C.-L., Aubry, C., Gutierrez, L. & Croue, J. P. 2013a Kinetic study of seawater reverse osmosis membrane fouling. Environmental Science & Technology 47 (19), 10884-10894.
- Khan, M. T., Manes, C.-L. d. O., Aubry, C. & Croué, J.-P. 2013b Source water quality shaping different fouling scenarios in a full-scale desalination plant at the Red Sea. Water Research 47 (2), 558-568.
- Khan, M. T., Busch, M., Molina, V. G., Emwas, A.-H., Aubry, C. & Croue, J.-P. 2014 How different is the composition of the fouling layer of wastewater reuse and seawater desalination RO membranes?. Water Research 59, 271-282.
- Khan, M. T., Hong, P.-Y., Nada, N. & Croue, J. P. 2015 Does chlorination of seawater reverse osmosis membranes control biofouling? Water Research 78, 84-97.
- Li, Z.-H., Li, Y., Gao, S.-X., Liang, W. & Guo, P.-S. 2019 Mitigated membrane fouling and enhanced removal of extracellular antibiotic resistance genes from wastewater effluent via an integrated pre-coagulation and microfiltration process. Water Research 159, 145-152.
- Pang, Y.-C., Xi, J.-Y., Xu, Y., Huo, Z.-Y. & Hu, H.-Y. 2016 Shifts of live bacterial community in secondary effluent by chlorine disinfection revealed by Miseq high-throughput sequencing combined with propidium monoazide treatment. Applied Microbiology and Biotechnology 100, 6435-6446.
- Reasoner, D. & Geldreich, E. 1985 A new medium for the enumeration and subculture of bacteria from potable water. Applied and Environmental Microbiology 49 (1), 1-7.
- Schneider, R. P., Ferreira, L. M., Binder, P. & Ramos, J. R. 2005 Analysis of foulant layer in all elements of an RO train. *Journal of Membrane Science* **261** (1–2), 152–162.

- Tan, Y.-J., Sun, L.-J., Li, B.-T., Zhao, X.-H., Yu, T., Ikuno, N., Ishii, K. & Hu, H.-Y. 2017 Fouling characteristics and fouling control of reverse osmosis membranes for desalination of dyeing wastewater with high chemical oxygen demand. Desalination 419, 1–7.
- Tang, F., Hu, H.-Y., Sun, L.-J., Wu, O.-Y., Jiang, Y.-M., Guan, Y.-T. & Huang, J.-J. 2014 Fouling of reverse osmosis membrane for municipal wastewater reclamation: autopsy results from a full-scale plant. Desalination 349, 73-79.
- Tang, F., Hu, H.-Y., Sun, L.-J., Sun, Y.-X., Shi, N. & Crittenden, J. C. 2016 Fouling characteristics of reverse osmosis membranes at different positions of a full-scale plant for municipal wastewater reclamation. Water Research 90, 329-336.
- Tong, X., Cui, Y., Wang, Y.-H., Bai, Y., Yu, T., Zhao, X.-H., Ikuno, N., Luo, H.-J., Hu, H.-Y. & Wu, Y.-H. 2020 Fouling properties of reverse osmosis membranes along the feed channel in an industrial-scale system for wastewater reclamation. Science of the Total Environment 713, 136673.
- Wang, Y.-H., Wu, Y.-H., Tong, X., Yu, T., Peng, L., Bai, Y., Zhao, X.-H., Huo, Z.-Y., Ikuno, N. & Hu, H.-Y. 2019 Chlorine disinfection significantly aggravated the biofouling of reverse osmosis membrane used for municipal wastewater reclamation. Water Research 154, 246-257.
- Xu, P., Bellona, C. & Drewes, J. E. 2010 Fouling of nanofiltration and reverse osmosis membranes during municipal wastewater reclamation: membrane autopsy results from pilot-scale investigations. Journal of Membrane Science **353** (1-2), 111-121.

- Xu, H.-B., de Koning, J. & Geng, Y. 2019 Reliability and efficiency of an advanced tertiary treatment process for wastewater reclamation. Journal of Water Reuse and Desalination 9 (4), 385-395
- Xu, L., Wei, C., Siddique, M. S. & Yu, W. 2020 Insight into the effect of in-situ galvanic micro-coagulation on membrane fouling mitigation treating surface water. Journal of Membrane Science 610, 118234.
- Yu, W.-Z., Qu, J.-H. & Gregory, J. 2015 Pre-coagulation on the submerged membrane fouling in nano-scale: effect of sedimentation process. Chemical Engineering Journal 262, 676-682.
- Yu, T., Li, G., Lin, W., Hu, H.-Y. & Lu, Y. 2016 Coagulation increased the growth potential of various species bacteria of the effluent of a MBR for the treatment of domestic wastewater, Environmental Science and Pollution Research 24 (6), 5126-5133.
- Yu, T., Meng, L., Zhao, Q.-B., Shi, Y., Hu, H.-Y. & Lu, Y. 2017 Effects of chemical cleaning on RO membrane inorganic, organic and microbial foulant removal in a full-scale plant for municipal wastewater reclamation. Water Research 113, 1-10.
- Zhang, J.-N., Lin, T. & Chen, W. 2017 Micro-flocculation/ sedimentation and ozonation for controlling ultrafiltration membrane fouling in recycling of activated carbon filter backwash water. Chemical Engineering Journal 325, 160-168.
- Zhao, X., Huang, H., Hu, H.-Y., Su, C., Zhao, J. & Liu, S.-M. 2014 Increase of microbial growth potential in municipal secondary effluent by coagulation. Chemosphere 109, 14-19.

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