Pre-coagulation on the submerged membrane fouling in nano-scale: Effect of sedimentation process

Wen-zheng Yu a,1, Jiu-hui Qu a,⇑, John Gregory b

a State Key Laboratory of Environmental Aquatic Chemistry, Research Center for Eco-Environmental Sciences, Chinese Academy of Sciences, Beijing 100085, China
b Department of Civil, Environmental and Geomatic Engineering, University College London (UCL), Gower Street, London WC1E 6BT, UK

HIGHLIGHTS
- Sedimentation process induced higher increase of TMP development.
- Flocs with larger primary particles were prone to be removed by sedimentation.
- Smaller primary particles after sedimentation caused higher density of cake layer.
- The zeta potential of flocs after sedimentation shifts in a negative direction.

ABSTRACT
Pre-coagulation and then flocculation with/without sedimentation before ultrafiltration (CSUF/CUF) was compared to explore the process for treating synthetic humic rich water and controlling membrane fouling. The sedimentation process caused larger flocs to be removed and zeta potential of the remaining flocs to be more negative. Also the fractal dimension of flocs became smaller after sedimentation process. All the data seemed to suggest that sedimentation process can improve the membrane fouling. On the contrary, the development of trans-membrane pressure (TMP) in CSUF surprisingly increased higher than that in CUF. The scanning electron microscope (SEM) images of cake layer on the surface of membrane showed that the size of nano-scale primary particles formed in coagulation process was heterogeneous. It also showed that the flocs formed by larger nano-scale primary particles were removed by sedimentation process, and the smaller nano-scale primary particles in CSUF maybe the crucial reason causing higher development of TMP. The density of cake layer on the CUF membrane surface with larger nano-scale particles was much lower than that by CSUF. Therefore, the characteristic of flocs, especially the size of nano-scale primary particles, may be the main factor inducing higher increase of TMP in CSUF, which should be explored in the future.

Crown Copyright © 2014 Published by Elsevier B.V. All rights reserved.

1. Introduction
Different pretreatments before ultrafiltration have been widely used as methods to mitigate membrane fouling in MF/UF membranes for drinking water treatment, such as coagulation, oxidation and adsorption. Combining oxidation and ultrafiltration can decrease the toxicity of the effluents and remove oxidation intermediates organic matter [1,2], and also can remove foulants from
amount of small micro-flocs decreased, and it was probably an flocs became smaller after flocs breakage and re-growth, the foul- of cake layer should be investigated to explore external membrane was smaller than about 3 nm and included about 85–90% of the [12] considered that when the colloidal fraction of material was pressible than for sweep floc conditions[10,11]. Howe and Clark tion produced lower hydraulic resistance and were less com- ing [20]. Park et al.[18] and Zhao et al.[21] reported that the flocs high fractal dimension, which could aggravate the membrane foul- previous work [22] also found that although the average size of membrane filtration, can significantly improve membrane fouling[5]. Adsorption by Powdered Activated Carbon (PAC) can also improve membrane fouling [5]. Pre-coagulation, as the most useful pretreatment before membrane filtration, can significantly improve membrane fouling [6–9]. Choi and Dempsey [10] found that in-line coagulation can improve hydraulic removal of filter cake. The severity of flux decline was greatly decreased when alum coagulant was added regardless the presence of calcium [7]. Flocs that were produced under charge neutralization conditions and acidic under-dose con- ditions produced lower hydraulic resistance and were less com- pressible than for sweep floc conditions [10,11]. Howe and Clark [12] considered that when the colloidal fraction of material was removed, the remaining dissolved organic matter (DOM), which was smaller than about 3 nm and included about 85–90% of the total DOM, caused very little fouling. Therefore, the characteristic of cake layer should be investigated to explore external membrane fouling. For pre-coagulation before membrane, perhaps the cake layer is the main factor causing membrane fouling. The characteristic of cake layer on the surface of membrane should be different by dif- ferent characteristic of flocs, such as by different coagulants [13], coagulant addition mode [14] or others. The characteristics, such as density, of cake layer on the surface of membrane are related to the structure of flocs [11,15,16]. Looser aggregates and fewer small colloidal particles cause lower membrane fouling [17–19]. Flocs of high humic water were small and regular in shape, with high fractal dimension, which could aggravate the membrane fouling [20]. Park et al. [18] and Zhao et al. [21] reported that the flocs with smaller size contributed to higher specific cake resistance and aggravated membrane fouling in coagulation–MF processes. Our previous work [22] also found that although the average size of flocs became smaller after flocs breakage and re-growth, the amount of small micro-flocs decreased, and it was probably an important factor contributing to lower increase of TMP. In traditional water treatment process, there was sedimentation process after coagulation and before sand filtration. But the perform- ance and mechanisms of the pretreatment with coagulation and then sedimentation process in membrane system has not been explored thoroughly, compared with only coagulation process. Usually, the additional sedimentation process demonstrates better performance than the direct filtration system without pretreat- ment, especially for a sudden increase of solid loading and severe fluctuation of source water properties [23]. Coagulation–with- out sedimentation–membrane systems was found can improve water flux and membrane fouling, and sedimentation can mitigate membrane fouling [6,24]. Control of cake permeabil- ity (and mass) via removing settleable flocs may decrease mem- brane fouling. Also different characteristic of flocs with/without sedimentation, including flocs size, fractal dimension and zeta potential, may cause different membrane fouling. Therefore, the different characteristic of cake layer on the surface of membrane formed by coagulated flocs with/without sedimentation process was explored to investigate the membrane fouling in this work. Although this work seems to be very straight- forward and sometimes determined by the raw water condition, unexpected results may provide some useful information, especially at nano-scale.

2. Materials and methods

2.1. Synthetic raw water and coagulant

Five grams of humic acid, sodium salt (HA, Aldrich, Cat: H1, 675-2), was dissolved in deionized (DI) water, with pH adjusted to 7.5 by 0.1 M HCl, and mixed by a magnetic stirrer for 24 h. The solution was diluted to 1 L in a measuring flask and was stored in the dark. The raw water used in this study was a mixture of tap water (Beijing, China) and humic acid solution at 5 mg/L and tap water was left for one night to release the residual chlorine. Synthetic raw water has been chosen here in order to simplify the study since natural surface water would have given reproducibility issues. The characteristics of raw water and tap water are listed in Table 1.

| Parameter | Raw water | Tap water |
|-----------|-----------|-----------|
| UV254 (cm⁻¹) | 0.131 ± 0.006 | 0.029 ± 0.004 |
| TOC (mg/L) | 4.78 ± 0.415 | 2.356 ± 0.234 |
| Turbidity (NTU) | 3.32 ± 0.16 | 0.42 ± 0.12 |
| Al (mg/L) | 0.068 ± 0.004 | 0.072 ± 0.005 |
| pH | 7.93 ± 0.06 | 7.90 ± 0.05 |

Note: for turbidity, UV254, DOC and pH, the number of measurements, n = 9; for residual Al, n = 5.

The solution was filtered through a 0.45 μm filter for 24 h. The filtrate was considered as synthetic raw water.

Aluminum sulfate hydrate (Al2(SO4)3·18H2O, >99%) ‘alum’ was used as a coagulant in this study. Stock alum solution was prepared at a concentration of 0.05 M in DI water.

2.2. Coagulation–UF processes

Coagulation–ultrafiltration (CUF) and coagulation–sedimentation–ultrafiltration (CSUF) were compared in the experiments. Pretreat- ment before UF processes of two systems were operated in parallel and a schematic illustration of two UF systems is shown in Fig. 1. Raw water was fed into a constant-level tank to maintain the water head for membrane tank. 0.1 mM Al (optimum alum dose was determined by removal of organic matter and zeta potential in jar test) was continuously added into the mix tank. The rapid mix speed was 200 rpm (180 s⁻¹) in the mix tank with hydraulic retention time (HRT) of 1 min, and then reduced to 50 rpm (23 s⁻¹) in the three floc- culation tanks with each HRT of 5 min. The relationship of mean velocity gradient (G) with the rotary speed is calculated following Hermawan et al. [25]. Coagulated water was either directly introduced into membrane tank (CUF) or first fed into the inclined sedimentation tank for flocs to settle (HRT = 15 min, 60°, length × diame- ter = 40 cm × 2 cm) and then influx into membrane tank (CSUF). The polyvinylidene fluoride (PVDF) hollow-fiber UF membrane module (Litree, China) with a nominal pore size of 0.01 μm and a surface area of 0.025 m² was submerged in the membrane tank. Permeate was continuously collected by a suction pump under the constant flux of 20 L/(m²·h⁻¹), operated in a cycle of 30 min filtration and 1 min backwash (40 L m⁻² h⁻¹), with a pressure gauge monitoring trans-membrane pressure (TMP). Air was supplied to each reactor at 100 L/h (air: water = 200:1) only at every backwash. The hydraulic retention time (HRT) was maintained at 0.5 h and sludge was discharged once a day.

2.3. Floc size and fractal dimension

A laser diffraction instrument (Malvern Mastersizer 2000, Mal- vern, U.K.) was used to measure the size and fractal dimension (Df) of flocs in the membrane tanks. The suspension attained from membrane tanks was monitored by drawing water through the optical unit of the Mastersizer and back into the jar by a peristaltic pump on the return tube with 5 mm internal diameter peristaltic pump tubing. Size measurements were taken every half minute for the duration of the jar test and logged onto a PC. This type of arrangement has previously been successfully applied in the analysis of coagulation flocs by Jarvis et al. [26]. Flocs were pumped through the system at a flow rate of 2.0 L/h. Preliminary
experimentation showed that flow rates above this value gave rise to floc breakage, while below this rate flocs settled in the tubing.

Mass fractals may be summarized by the relationship between their mass $M$, a characteristic measure of size $L$, and the mass fractal dimension $D_f$: 

$$M \propto L^{D_f}$$  \hspace{1cm} (1)

During small-angle light scattering (SALLS), a light beam is passed through a sample. The particles in the sample scatter light proportionally to their size and at a constant angle independent of which part of the particle is hit by the beam. Small particles scatter light at high angles, while large particles scatter at lower angles. The Malvern Mastersizer has an array of photosensitive detectors at different angles between 0.01° and 40.6° that detect the light scattered by the sample. The determination of fractal dimension by use of SALLS has been well illustrated [27–29], so only a brief overview is given below.

The total scattered light intensity $I$ is a function of the scattering vector $Q$, where $Q$ is the difference between the incident and scattered wave vectors of the radiation beam in the medium [29], which is given by (2).

$$Q = \frac{4\pi n \sin (\delta/2)}{\lambda}$$  \hspace{1cm} (2)

where $n$ is the refractive index of the suspending medium, $\delta$ is the scattering angle, and $\lambda$ is the wavelength of the radiation in a vacuum. For independently scattering aggregates, $I$ is related to $Q$ and the fractal dimension by $D_f$ (3):

$$I \propto Q^{D_f}$$  \hspace{1cm} (3)

A confirmation of the power relationship by Spicer et al. [30] is to plot $I$ against $Q$ on a log-log scale. A power law relationship exists if this yields a straight line, the slope of which is used to give $D_f$. The relationship only holds when the length of investigation is much larger than the primary particles and much smaller than the floc aggregates.

### 2.4. Other analytical methods

Zeta potential distribution of flocs in both two membrane tanks was measured by a zeta meter (Zetasizer nano series, Malvern, UK). The concentration of suspended solids (SS) in synthetic raw water and coagulated suspensions was measured after 0.45 μm membrane filtration, weighting the difference before and after dried at 105 °C. Turbidity was determined by a WTW TURB555iR turbidimeter. UV absorbance (after 0.45 μm) at the wavelength of 254 nm (UV254) was determined by an ultraviolet/visible spectrometer (U-3010 Hitachi High-Technologies Co., Japan). Dissolved organic carbon (DOC) of water samples (after 0.45 μm membrane filtration) was determined with a total organic carbon (TOC) analyzer (TOC-VCPH, Shimadzu, Japan). Residual aluminum after 0.45 μm membrane was measured by inductivity coupled plasma optical emission spectrometer (ICP-OES,710, Agilent Technologies, USA).

The fouled membrane fibers were cut from the two membrane modules, and the foulant layer attached on the membrane surface was retained. The fouled membrane samples were then platinum-coated by a sputter and observed under scanning electron microscopy (SEM; JSM7401F, JEDL, Japan).

### 3. Results and discussion

#### 3.1. Zeta potential distribution of flocs

As the flocs in CUF were larger than the upper size limit for the zeta meter, zeta potential distribution of flocs in CUF and CSUF cannot be measured accurately. The zeta potential of flocs will not be changed during coagulation process, even after breakage [31]. Therefore, zeta potential distribution of alum-humic floc in jar test after rapid mixing and settling for 30 min after flocculation was used for CUF and CSUF systems, respectively.

The main difference was that the zeta potential distribution of flocs after settling was displaced in a negative direction compared to that after rapid mixing (Fig. 2), which was the same as the result of McCurdy et al. [32]. A shift in the charge distribution from suspended floc to coagulated flocs may indicate suspended particles in CSUF were different from the coagulated flocs in CUF, which was supported by the fractal dimension ($D_f$), size of flocs and primary nano-scale particles discussed later. The sedimentation process in CSUF treatments resulted in particles being more suitable for filtration since less flocs remained before filtration and the charge of particles became more negative, which seemed more beneficial for controlling membrane fouling as the membrane was negatively charged (around −10 mV, provided by membrane company). But comparing TMP development later, more negative charge of flocs...
in CSUF caused higher membrane fouling, and it could not be concluded that the membrane fouling was determined by the zeta potential.

3.2. Water quality of effluents of coagulation–UF systems

The removal efficiencies of pollutants (turbidity and dissolved organic matter) by CSUF and CUF at the dose of 0.1 mM alum (calculated as Al\(^{3+}\)) were listed in Table 2. Turbidity was reduced from initial 3.32 ± 0.16 NTU in raw water to a level less than 0.1 NTU in both effluents. Furthermore, there were no significant differences of average removal of DOC and UV\(_{254}\) between the two systems during the operation period, suggesting that sedimentation process did not give any increase in organic removals. These results implied that the removal efficiency of organic matter was independent on the sedimentation process. The dissolved organic matter (DOM), as well as same residual Al in both membrane systems, was not the factors determining the membrane fouling, which was the same as the result of Howe and Clark [12]. It meant that DOM (or Al-humic complex) after coagulation and residual Al would not cause membrane fouling. As shown in Table 2, the only difference was the turbidity in the CUF and CSUF tank, which was a little higher in CUF tank. The higher turbidity in CUF also seemed to support the development of TMP in CUF should be higher than the one in CSUF.

3.3. Characteristic of flocs in CUF and CSUF

Floc characteristics, such as floc size, fractal dimension (\(D_f\)) and amount, are very important for continuous membrane filtration. Thus, the particle (floc) size distribution (PSD) and \(D_f\) of flocs in CUF membrane tank (without sedimentation) and CSUF membrane tank (after sedimentation) were explored (Fig. 3a). PSD in CUF membrane tank was mainly between 5 and 250 \(\mu\text{m}\), and there were many flocs smaller than 2 \(\mu\text{m}\), which may influence the characteristic of cake layer on the surface of membrane. While in CUF membrane tank, PSD was mainly between 60 and 500 \(\mu\text{m}\), and there were small flocs but larger than 15 \(\mu\text{m}\). The result meant that larger flocs were removed in sedimentation process, which was supported by the SS concentration, and it was higher in CUF tank than in CSUF tank. The cake layer formed by these two different characteristic of flocs should be different, as the characteristic of cake layer is mainly determined by the smaller flocs [19]. From Fig. 3b, the fractal dimension of flocs in CUF membrane tank was near 2.65, while the one in CSUF membrane tank was only 2.49. Normally, the higher the fractal dimension of flocs, the higher density of cake layer formed by flocs. But from the results of Fig. 3b, higher fractal dimension (2.65) caused lower TMP development.

Howe and Clark [12] considered that when the colloidal fraction of material was removed, the remaining dissolved organic matter (DOM), which was smaller than about 3 nm and included about 85–90% of the total DOM, caused very little fouling. The results meant that the nano-scale humic-Al particles retained on the surface of UF membrane would not cause inner membrane fouling.

### Table 2

| Parameter      | Raw water | CUF tank | CSUF tank | CUF effluent | CSUF effluent |
|----------------|-----------|----------|-----------|--------------|---------------|
| UV\(_{254}\)  | 0.131 ± 0.006 | 0.032 ± 0.002 | 0.031 ± 0.002 | 0.031 ± 0.003 | 0.030 ± 0.002 |
| DOC (mg/L)     | 4.282 ± 0.415 | 2.232 ± 0.185 | 2.132 ± 0.302 | 2.042 ± 0.234 | 2.088 ± 0.287 |
| Turbidity (NTU)| 3.32 ± 0.16 | 2.53 ± 0.18 | 1.85 ± 0.21 | 0.05 ± 0.02 | 0.06 ± 0.03 |
| Al (mg/L)      | 0.068 ± 0.004 | 0.113 ± 0.002 | 0.108 ± 0.002 | 0.083 ± 0.001 | 0.078 ± 0.003 |
| pH             | 7.93 ± 0.06 | 7.38 ± 0.04 | 7.34 ± 0.04 | 7.40 ± 0.05 | 7.37 ± 0.03 |

Note: for turbidity, UV\(_{254}\), DOC and pH, the number of measurements, \(n\) = 9; for residual Al, \(n\) = 5.

![Fig. 2. Zeta potential distribution for floc in jar test.](image)

![Fig. 3. Characteristic of flocs in CUF and CSUF: (a) size distribution of flocs, (b) fractal dimension of flocs.](image)
but just external fouling. Because of the greater proportion of small particles in the CSUF system, these particles were more prone to form high density cake layer on the surface of membrane. Thus, size of flocs, especially small flocs, may be one of the main factors determining the density of the cake layer. Also the size of nano-scale primary particles composing flocs was explored to further confirm the result later.

3.4. TMP developments of CUF and CSUF

In these experiments, the TMP was used as the indicator of membrane fouling by maintaining the constant value of flux in CUF and CSUF systems, and it increased with operation time (Fig. 4), and membrane filtration without pretreatment is also shown. Neither physical cleaning nor chemical cleaning was carried out during 32 days of operation. The TMP increased with time from the initial 11 kPa. The membrane fouling increased significantly if there was no pretreatment before membrane filtration, which meant that humic acid can induce serious membrane fouling if there was no pretreatment. While for the two pretreatment systems, the TMP increased quickly for both systems with a similar trend within the beginning phase (1 day), from 11 kPa to 13 kPa.

Then the TMP development of both CUF and CSUF membrane tanks exhibited a slow increase for nearly 20 days followed by a relative rapid one. However, there was a distinct difference of 1.0 kPa between them after 3 days. At the end of the process (32 days),

Fig. 4. Comparison of TMP development in CUF and CSUF.

Fig. 5. SEM image of the cake layer on the surface of membrane: (a) ×20,000 and (b) ×50,000-fold fouled CUF membrane, (c) ×20,000 and (d) ×50,000-fold fouled CSUF membrane, (e) size distribution of nano-particles in CUF and CSUF cake layer.
the TMP for CUF increased to 24.5 kPa, while that of CSUF increased to 33 kPa. It seemed that removal of large flocs by sedimentation process did not improve the membrane fouling. On the contrary, the sedimentation process caused higher membrane fouling.

External fouling and internal fouling were explored here. The cake layer (external foulants) on membrane surface was carefully washed away with high pressure tap water at the end of the operation period, and then the filtration process was restored again using DI water. The TMP of both membrane systems were reduced to 11 kPa, which was the same as that of the new membrane. It indicated that the reversible fouling dominated membrane fouling mechanism in both systems. Little membrane fouling was determined by the residual organic compounds after coagulation, as NOM before and after membrane filtration was nearly the same.

Above all, it should be noted that the sedimentation process caused higher external membrane fouling (TMP). Therefore, the cake layer at nano-scale on the surface of membrane was investigated.

3.5. Microscopic observations of membrane surface

In order to further investigate the membrane fouling mechanism, the cake layer on the surface of membrane was investigated. Fig. 5 illustrates membrane surface images obtained from the SEM analysis, showing the fouled membrane surface with cake layer in CUF without sedimentation process and the one in CSUF with sedimentation process. After adding alum into water, it hydrolyzed immediately in less than one second [33], and then formed nano-scale primary particles. After that, alum-humic complex was adsorbed on the primary particles and then aggregate into large flocs after flocculation process. The size of nano-scale primary particles on the two membranes was significantly different. The primary particles of cake layer on the surface of CUF membrane, are much larger than those on CSUF membrane (Fig. 5). Therefore, it can be speculated that the size of nano-scale primary particles formed after Al hydrolysate is not homogeneous (Fig. 6), and then these primary particles aggregate into large flocs by coagulation process. Large flocs can be formed by the both small and large nano-scale primary particles, but the ones formed by larger primary particles seems to be removed easier by sedimentation process, while flocs with smaller primary particles were difficult to settle and became part of cake layer in CSUF. The lower fractal dimension of flocs in CSUF than the one in CUF also support its low settling ability.

From Fig. 5, the density of cake layer seems to be low and there are relative larger pores on the surface of CUF membrane (Fig. 5a and b). On the contrary, there were smaller pores in the cake layer on the surface of CSUF, and the density was relatively higher (Fig. 5c and d). The statistic size of nano-particles in the two membrane cake layers by SmileView software clearly showed that the size of nano particles in CUF cake layer are larger than that in CSUF cake layer (Fig. 5e). The density of cake layer increased since the size of primary particles decreased as the pore volume of cake layer increased in CSUF cake layer. Cake layer permeability decreases with decreasing particle size. Also the number of connection points between primary particles decreased as the size of nano-scale primary particles increased, and it was more difficult for smaller nano-scale particles to be removed by backwash and aeration (backwash and aeration did not change the size of nano-scale primary particles). Fig. 6 can clearly show the mechanism of different membrane fouling by two pre-treatments. The smaller flocs and smaller nano-scale primary particles may be the main reason causing the higher membrane fouling in CSUF system, which should be investigated in the future.

4. Conclusions

The main conclusions of this work are:

1. Removal of large flocs by sedimentation process in CSUF did not improve the membrane fouling well, but caused higher increase of TMP development than the one in CUF without sedimentation.

2. The size of nano-scale primary particles formed in coagulation process was heterogeneous. Flocs formed by larger nano-scale primary particles were prone to be removed by sedimentation process in CSUF system, which induced smaller nano-scale primary particles attached on the surface of membrane in CSUF tank. Smaller flocs and smaller nano-scale primary particles in CSUF membrane tank caused higher density of cake layer than the one in CUF tank.

3. The zeta potential distribution of flocs after sedimentation process shifts in a negative direction compared to the one without sedimentation process. More negative charged particles in CSUF caused higher increase of TMP development, which may not determine the membrane fouling.

4. Fractal dimension is not the factor determining the increase of TMP development here, as higher fractal dimension (2.65) of flocs in CUF caused lower increase of TMP compared with lower fractal dimension (2.49) of flocs in CSUF.

Acknowledgements

This work was also supported by National Natural Science Foundation of China (Grants 51138008 and 51108444). Dr Yu’s research was also supported by a Marie Curie International Incoming Fellowship (FP7-PEOPLE-2012-IIF-328867) within the 7th European Community Framework Programme.

References

[1] D. Arsene, C.P. Musteret, C. Catrinescu, P. Apopei, G. Barjoveanu, C. Teodosiu, Combined oxidation and ultrafiltration processes for the removal of priority organic pollutants from wastewaters, Environ. Eng. Manage. J. 10 (2011) 1967–1976.

[2] M.D. Murcia, M. Gomez, E. Gomez, A. Bodalo, J.L. Gomez, A.M. Hidalgo, Assessing combination treatment, enzymatic oxidation and ultrafiltration in a membrane bioreactor, for 4-chlorophenol removal: experimental and modeling, J. Membr. Sci. 342 (2009) 198–207.

[3] T. Liu, Z.L. Chen, W.Z. Yu, S.J. You, Characterization of organic membrane foulants in a submerged membrane bioreactor with pre-ozonation using
three-dimensional excitation–emission matrix fluorescence spectroscopy, Water Res. 45 (2011) 2111–2121.

[4] H. Fang, D.D. Sun, M. Wu, W. Phay, J.H. Tay, Removal of humic acid foulant from ultrafiltration membrane surface using photocatalytic oxidation process, Water Sci. Technol. 51 (2005) 373–380.

[5] J. Haberkamp, A.S. Ruhl, M. Ernst, M. Jekel, Impact of coagulation and adsorption on DOC fractions of secondary effluent and resulting fouling behaviour in ultrafiltration, Water Res. 41 (2007) 3794–3802.

[6] K. Konieczny, M. Raja, M. Bodzek, A. Kwiecinska, Water treatment using hybrid method of coagulation and low-pressure membrane filtration, Environ. Prot. Eng. 35 (2009) 5–22.

[7] K. Listiarini, D.D. Sun, J.O. Leckie, Organic fouling of nanofiltration membranes: evaluating the effects of humic acid, calcium, alum coagulant and their combinations on the specific cake resistance, J. Membr. Sci. 332 (2009) 56–62.

[8] R.H. Peiris, M. Jaklewicz, H. Budman, R.L. Legge, C. Moresoli, Assessing the role of feed water constituents in irreversible membrane fouling of pilot-scale ultrafiltration drinking water treatment systems, Water Res. 47 (2013) 3364–3374.

[9] H.E. Wray, R.C. Andrews, P.R. Berube, Ultrafiltration organic fouling control: comparison of air-sparging and coagulation, J. Am. Water Works Assoc. 106 (2014) E76–E85.

[10] K.Y.J. Choi, B.A. Dempsey, In-line coagulation with low-pressure membrane filtration, Water Res. 38 (2004) 4271–4281.

[11] J.D. Lee, S.H. Lee, M.H. Jo, P.K. Park, C.H. Lee, J.W. Kwak, Effect of coagulation conditions on membrane filtration characteristics in coagulation–microfiltration process for water treatment, Environ. Sci. Technol. 34 (2000) 3780–3788.

[12] K.J. Howe, M.M. Clark, Fouling of microfiltration and ultrafiltration membranes by natural waters, Environ. Sci. Technol. 36 (2002) 3571–3576.

[13] J. Wang, J. Guan, S.R. Santivong, T.D. Waite, Characterization of floc size and structure under different monomer and polymer coagulants on microfiltration membrane fouling, J. Membr. Sci. 321 (2008) 132–138.

[14] T. Liu, Z.L. Chen, W.Z. Yu, J.M. Shen, J. Gregory, Effect of two-stage coagulant addition on coagulation–ultrafiltration process for treatment of humic-rich water, Water Res. 45 (2011) 4260–4268.

[15] D. Antelmi, B. Cabane, M. Meireles, P. Aimar, Cake collapse in pressure filtration, Langmuir 17 (2001) 7137–7144.

[16] P.K. Park, C.H. Lee, S. Lee, Determination of cake porosity using image analysis in a coagulation–microfiltration system, J. Membr. Sci. 293 (2007) 66–72.

[17] M.H. Cho, C.H. Lee, S. Lee, Effect of flocculation conditions on membrane permeability in coagulation–microfiltration, Desalination 191 (2006) 386–396.

[18] P.K. Park, C.H. Lee, S. Lee, Permeability of collapsed cakes formed by deposition of fractal aggregates upon membrane filtration, Environ. Sci. Technol. 40 (2006) 2699–2705.