Development of an efficient approach for separating bubbles and flocs in a submerged membrane ultrafiltration process
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ABSTRACT
This work was carried out to investigate the impact of air scouring and membrane configuration on ultrafiltration (UF) performance in a hybrid process of coagulation-UF. To eliminate the adverse impacts of air scouring, the concept of separating flocs in the submerged membrane module was proposed. The performance of three types of floc separation devices, including a cylinder module, a module without a floc separation device and a stacked inclined plate module, was compared. In the coagulation-UF process (i.e., without air scouring), the average transmembrane pressure (TMP) growth rates of the cylinder module, the module without a floc separation device and the inclined plate module were 0.58, 0.76 and 1.38 kPa/h, respectively, indicating lighter membrane fouling of the former two membrane configurations. In the coagulation-air scouring-UF process (with an air scouring rate of 35 mL/min), the stacked inclined plate module showed better effluent water quality (lower UV254) and floc integrity (the floc size was about twice that of the other two configurations). Further, the optimization of the air scouring rate was conducted when the inclined plate module was used, and the optimal value was determined to be 60 mL/min for air rates ranging from 0 to 100 mL/min.

Key words | air scouring, floc separation, membrane configuration, submerged membrane process, ultrafiltration

INTRODUCTION
Application of ultrafiltration (UF) in municipal water treatment has undergone rapid development in recent decades. However, membrane fouling is always the main obstacle for wider implementation of UF (Gao et al. 2011). To minimize UF membrane fouling, various approaches including integration with pretreatment, optimizing the operating conditions and modifying the membrane modules have been investigated (Li et al. 1998). Coagulation has been the most successful pretreatment method for the fouling control of UF membranes in full-scale water treatment (Huang et al. 2009a). Coagulation enables the formation of a porous cake layer and limits pore blockage by aggregating the particles and organic matter (Barbot et al. 2008). More irregular and branched aggregates with higher fractal dimension were produced by a sweep-floc mechanism, but the formed flocs were fragile and easily broken, which might cause the release of small particles and natural organic matter (NOM) (Howe et al. 2006). Coagulation performance and its impact on membrane fouling reduction were affected by many factors, for example, the type and dosage of coagulant, pH, coagulant viscosity and coagulant dosing method (Wang et al. 2016; Shen et al. 2017). Howe & Clark (2006)
systematically evaluated the impact of coagulation pretreatment on the membrane filtration performance.

Regarding the hydrodynamic disturbance methods, air scouring is an effective and low-cost approach to alleviate membrane fouling (Li et al. 1997). The effect of air scouring on the membrane performance is complex and related to the bubble characteristics (Cui et al. 2003), bubbling mode (continuous or intermittent air sparging) (Tian et al. 2010) and membrane module configuration (Liu et al. 2014). Ye et al. (2006) found that aeration from an off-center position resulted in superior membrane performance than the center position because of the reduction of ‘dead zones’, and multiple small bubbles were more effective than fewer large bubbles at the same gas flow rate. Pradhan et al. (2012) observed that a higher air flow rate for membrane scouring resulted in lower membrane fouling. Cabassud et al. (2001) discovered that flux enhancement was attributed to fluid mixing and turbulence near the membrane surface generated by bubbles. Bubbles exert influence mainly in two ways: enhancing the shear stress of the membrane surface and affecting the status of the particulate pollutant, which includes altering the particle movement and particle size (Cui et al. 2003; Qaisrani & Samhaber 2011).

In the study of UF membrane fouling control, coagulation-UF or air sparging-UF were two widely used processes. Liu et al. (2014) combined the three processes of coagulation, air sparging and UF to research the interaction of particles, bubbles and membranes. In their research, gas sparging induced shear rate limited concentration polarization but caused floc breakage, leading to the release of small colloids and NOM, which could accelerate membrane fouling. It seemed that some undesirable effects would occur when the two membrane fouling control methods worked together. Based on this observation, we were wondering if we could separate the flocs from the membrane module under the bubbling condition, as the adverse effects of the combined process would be eliminated and the membrane performance would be promoted.

Referring to the principle of the inclined plate sedimentation tank, a new type of floc separation device made up of stacked truncated cone-shaped inclined plates was developed and used in the membrane module to separate bubbles and flocs in a hybrid process of coagulation-UF with air scouring applied. This study was carried out with three objectives. (a) To compare the performance of the newly-developed inclined plate module and two other types of floc separation membrane configurations (discussed in the following section) under different air scouring conditions. (b) To study the effectiveness of the three membrane configurations in floc sedimentation and their impacts on membrane fouling control and UF effluent water quality. The hydraulic performance and structure of the flocs in the UF reactor were also studied under aeration conditions. (c) The optimization of the air scouring rate was also conducted when the inclined plate module was used.

**MATERIALS AND METHODS**

**Feed water**

Raw water was drawn from Mingyuan Lake (located in Sichuan University) every day in this study. Al2(SO4)3·18 H2O (Sigma Aldrich, analytical grade) was used as a coagulant. To determine the optimal coagulant dosage, a jar test was performed in a flocculator (ZR4-6, Zhongrun, China). Coagulation was conducted in two stages. First, a rapid stirring speed of 500 r/min was maintained for 1.5 min; then, the rotation speed was reduced to 60 r/min for 22 min to allow floc growth. After stirring, the coagulated water was rested for 30 min, and the turbidity and UV254 of the supernatant were measured. The optimal dosage was determined as 0.21 mmol/L (in Al), as shown in Figure S1 (supplementary information, available with the online version of this paper).

**Three membrane configurations for floc separation**

Figure 1 presents the configurations of the three membrane setups for floc separation. As shown in Figure 1(a), there was no auxiliary sedimentation device outside the membrane. This is a common device in previous research (Yu et al. 2011; Xia et al. 2015). As illustrated in Figure 1(b), Liu et al. (2014) made some improvements: a cylindrical sleeve was added inside the module as a floc collection device. In this type of membrane module, hollow fibers were mounted between the reactor shell and the cylindrical sleeve, as shown in Figure S2 (available online). Flocs that enter the reactor move upward under the drag of the bubbles until
reaching the top edge of the sleeve. After surmounting the top edge, flocs start to fall down toward the bottom of the sleeve under gravity. There was a sludge outlet at the bottom of the sleeve, from which the flocs and particles that settled were discharged. The floc separation efficiency of this device is low because precipitation occurs only in the top region of the sleeve; thus, further improvement of the membrane module is necessary.

Referring to the configuration of the inclined plate sedimentation tank, we developed a novel floc separation device composed of stacked truncated cone-shaped inclined plates. A 3D model diagram of the inclined plate floc separation device is shown in Figure 1(c). The entire structure, with a total height of 230 mm, comprises 20 pieces of truncated cone-shaped inclined plates, which form the working parts of the device. The top diameter and bottom diameter of a single piece of the inclined plate are 22 and 32.4 mm, respectively, and the inclined angle is 60°. The working principle of the inclined plate module is illustrated in Figure 1(c). Coagulated water enters the membrane tank through the water inlet. The flocs and particles then move upward under the drag force generated by the bubbles, which are produced by a micro-porous aeration diffusor installed on the bottom. At the same time, the upward-moving flocs and particles slide down the slope of the inclined plates under gravity. The inclined plates have two functions: providing large surfaces for floc settling and serving as isolation media for the flocs and hollow fibers. The flocs that slide down settle to the bottom and are then discharged through the sludge outlet. To make the device, a modeling software SketchUp was used to draw the three-dimensional model and a 3D printer (SIGAO 400, Chengdu Creative Three Dimensional Technology Co., Ltd, China) was used to print the model using polylactic acid (PLA). A dissolution test indicated that the dissolution of PLA into water could be ignored.

It is worth mentioning that hollow fibers and an aeration diffusor were placed inside the cylinder when membrane fouling tests were performed with the cylinder module (Figure 1(b)) (differing from the case in Figure S2, where hollow fibers and an aeration diffusor were placed outside the cylinder). We made such a change because the settling area outside the cylinder is larger than that inside the cylinder. Thus, a larger floc settling area was provided, and a better floc separation was expected. For convenience, the membrane configuration without a floc separation device (Figure 1(a)), the membrane configuration with a cylinder floc separation device (Figure 1(b)) and the membrane configuration with inclined plates (Figure 1(c)) are referred as MC1, MC2 and MC3, respectively.

**Experimental procedures and membrane performance evaluation**

The outside-in polyvinylidene fluoride (PVDF) hollow fibers (Litree Purifying Technology Co., Ltd, Hainan, China) with a nominal pore size of 0.02 μm were installed vertically, and the lower end of the fibers were not fixed. The inner and external diameters of the hollow fibers are 1.0 and
1.8 mm, respectively. There are 16 hollow fibers in each module and the effective length of the hollow fibers is 20 cm, with a total active membrane area of 0.018 m². The water contact angle of the new membrane is 42.2 ± 2.6°.

A bench-scale setup of the coagulation-bubbling-UF process is shown in Figure 2. The coagulation stage was completed in three reaction tanks. In the rapid mixing tank, the rotation speed was set at 300 r/min, and the hydraulic retention time was 1.3 min. The rotation speed and hydraulic retention time were 60 r/min and 11 min, respectively, in both flocculation tanks. In the submerged UF module, one of the flocc separation devices was placed on the periphery of the hollow fibers when membrane fouling tests were carried out. Water was filtered under the suction of a peristaltic pump (BT100-2 J, Longer Pump, China). The transmembrane pressure (TMP) was monitored by a vacuum gauge and a pressure sensor simultaneously. Each UF test lasted 7.5 h and was operated under constant flux mode with a value of 20 L/(m²·h). Membrane performances under different air scour rates were explored and the parameters of the air bubbles applied in our study are presented in Table S3 (available online).

UF membrane fouling was determined by the TMP buildup and UMFI (unified membrane fouling index), as proposed by Huang et al. (2008, 2009b):

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\frac{1}{J'_S} = 1 + (\text{UMFI})V_S
\]

where \(V_S\) (L/m²) is the permeate volume of the unit membrane area and \(J'_S\) is the normalized membrane specific flux. For constant flux mode, \(J'_S = \frac{\text{TMP}_0}{\text{TMP}}, \text{TMP}_0\) and \(\text{TMP}\) (kPa) are the TMP of the new membrane and the TMPs after the membrane was fouled (Chang et al. 2016).

**Analytical methods**

The turbidity was measured with a HACH 2100P (HACH company, Loveland, Colorado, USA) turbidity meter. UV₂₅₄ was measured with a spectrophotometer (Aquamate UV-VIS, Thermo Fisher Scientific, USA) after the lake water was pre-filtered with a 0.22 μm membrane.

A Zetasizer Nano ZS (Malvern Instruments Ltd, Malvern, UK) was used to measure the size of the particles and flocs in the UF column. Water samples were taken from the UF column after membrane fouling tests were carried out for 2 h. For each sample, measurements were performed three times, and an average result was calculated. A scanning electron microscope (SEM) (JSM-7500F, JEOL Ltd, Japan) was used to obtain surface images of the clean and fouled membranes. Before taking pictures, the hollow fibers were sputter coated (60 s with an operating current of 10 mA) with gold using a sputter coater (Q150R ES, Quorum, UK).

Tracer tests were conducted to analyze the fluid dynamics with rhodamine B dye. The tracer was introduced at the UF reactor inlet continuously (step input), and the
response was measured at the outlet. The influent level of rhodamine B is 1 g/L for all the tracer tests and the tracer concentration at the outlet was expressed as normalized tracer concentration \( C/C_0 \) (\( C \) and \( C_0 \) represent the tracer concentration in the effluent and the influent, respectively).

**RESULTS AND DISCUSSION**

**Feed water characteristics**

The water quality was monitored continuously during the experiment and the results are as follows: temperature, 23.4-29.9 °C; pH, 8.21-8.95; turbidity, 3.22–6.98 NTU; SS, 4.99–9.35 mg/L; UV\(_{254}\) 0.098–0.118 cm\(^{-1}\). After coagulation, turbidity and UV\(_{254}\) decreased to 1.47–2.39 NTU and 0.059–0.068 cm\(^{-1}\), respectively. The influent turbidity and UV\(_{254}\) in each experiment and the water quality of bulk water (after coagulation) in each module are presented in Table S4 (available with the online version of this paper).

**Role of membrane configurations in the coagulation-UF process**

In the coagulation-UF process (i.e., no air scouring was applied), the difference between the turbidity of the effluent water of the three configurations was within 0.03 NTU, and the UV\(_{254}\) values were all in the vicinity of 0.06 cm\(^{-1}\). The results indicated that none of the three membrane configurations showed a significant advantage over the others in terms of contamination removal in the no-air scouring condition.

Figure 3 shows the TMP development and UMFI value of the three membrane configurations in the coagulation-UF process. Figure 3(a) illustrates that MC1 and MC2 showed relatively slow TMP growth in the no-bubbling condition, and the growth rates of the two configurations were 0.76 and 0.58 kPa/h, respectively, which were lower than that of MC3 (1.38 kPa/h). The UMFI was calculated to eliminate the impact of the initial TMP difference under different control conditions (as shown in Figure 3(a)) and to quantify the membrane fouling potential in the entire filtration circle (Huang et al. 2008). As shown in Figure 3(b), the UMFI values of MC1 and MC2 were considerably lower than that of MC3. The lowest UMFI appeared in MC2, consistent with the result based on the TMP growth, which indicated a better membrane performance when the cylinder floc separation device was used in the no-air scouring condition. The better performance of MC1 and MC2 under the no-air scouring condition can be explained from the perspective of floc sedimentation. When there was no gas bubbling, flocs that entered the UF column settled to the bottom under gravity. In MC1 and MC2, the flocs could settle without any obstruction and achieve good separation from the hollow fibers. However, in MC3, considering the structure of the stacked inclined plates, these inclined plates actually served as barriers, which hindered the free settling of the flocs. In the membrane fouling tests, many foulants were blocked between the inclined plates and other portions deposited on the membrane surface, which enhanced the adsorption

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**Figure 3** | Membrane fouling comparison of the three membrane configurations in the coagulation-UF process: (a) TMP development trend and (b) UMFI value.
of the particles on the membranes. Thus, membrane fouling was accelerated in MC3.

Role of membrane configurations in the coagulation-air scouring-UF process

When 35 mL/min of air scouring was applied, the turbidity of the three effluent water conditions fluctuated slightly. However, there was a notable decline in the UV254 value of the filtered water that flowed out from the inclined plate membrane (the UV254 values for MC1, MC2 and MC3 are 0.056, 0.056 and 0.042 cm$^{-1}$, respectively). In water quality analyses, UV254 (ultraviolet absorbance at 254 nm) is often identified as a substitutive measure for the DOC (Matilainen et al. 2014). The low UV254 value of the effluent water may be related to the good integrity of the flocs in the module. This meant that the stacked inclined plates provided a good hydraulic condition in the module, under which the formed flocs were not easily broken. Thus, less NOM was released from broken flocs than with the other two membrane configurations. The explanation of the floc integrity is further demonstrated through particle size analysis below. In sedimentation theory, the overflow rate is an important parameter affecting the particle precipitation efficiency. It can be obtained by dividing the process flow rate by the effective sedimentation area (further explanation about overflow rate is presented in the supplementary information, S5, available online). For a specific sedimentation basin and particles, a smaller overflow rate will result in better particle removal (Crittenden et al. 2012). The overflow rate of MC3 (0.02 m$^3$/(m$^2$·h)) is much smaller than that of MC1 and MC2 (both are 0.29 m$^3$/(m$^2$·h)), which means MC3 has the potential to remove a larger fraction of particles. This could also produce a better effluent water quality.

When 35 mL/min of air scouring was applied, the TMP development was similar to that in the coagulation-UF process. In this case, the TMPs of MC1 and MC2 also showed a relatively slow growth rate compared to MC3, as shown in Figure 4(a). The tracer tests were carried out to reveal the mechanism of TMP growth and the results are shown in Figure 5(a). The tracer curves under different conditions indicated that MC1 and MC2 had a shorter hydraulic retention time (rhodamine B could be quickly detected at the outlet) compared with MC3. Considering the complex structure of the stacked truncated cone-shaped inclined plates of MC3 (Figure 1(c)), these inclined plates with a large vertical projection area caused large hydraulic resistance when water flowed in the vertical direction. The flow serpentinized in the membrane module (Crittenden et al. 2012). Thus, the interaction between the hollow fibers and flocs was enhanced due to the disturbance of air bubbles. The deterioration of the membrane performance caused by the strong interaction between the hollow fibers and flocs has also been reported by Liu et al. (2014). In comparison, the vertical hydraulic resistances in MC1 and MC2 were small when water flowed in the channels of the membrane reactors. The lateral flow in these two membrane reactors was weak.

Figure 4 Membrane fouling comparison of the three membrane configurations in the coagulation-air scouring-UF process (with an air scouring rate of 35 mL/min): (a) TMP development trend and (b) UMFI value.
The effect of water flow on the membrane surface was mainly shear stress, which could effectively inhibit membrane fouling. This might be the reason why the TMP grew relatively slowly when using MC1 and MC2. MC3 still maintained a high UMFI value in the coagulation-air scouring-UF process (Figure 4(b)). The analysis of the TMP and UMFI indicates that the import of the inclined plate floc separation device accelerated membrane fouling, whereas the cylinder configuration showed similar membrane performance to the membrane without the auxiliary sedimentation device.

Comparing Figures 3(a) and 4(a), in the coagulation-UF process, the TMP continued rising at a high speed even at the end of the membrane fouling test. While in the coagulation-air scouring-UF process, the TMP development of the three modules exhibited a two-stage trend: rapid development was observed in the first few hours, and then the TMP slowed down after approximately 5 h. The different experimental results verified the effectiveness of air bubbling in improving the membrane performance in the long-term operation. This observation was consistent with the result reported by Liu et al. (2014).

Figure 5(b) illustrates clear differences between the floc size distributions for the three membrane configurations. The average floc size in MC3 (approximately 7,400 nm) is considerably larger than that in the other two configurations (both are approximately 3,500 nm for MC1 and MC2). This result proved the good integrity of the flocs in the inclined plate configuration, which supports the hypothesis outlined above. In addition, there was a portion of the flocs in the size range of 10–20 nm, which was similar to the pore size of the hollow fibers. Foultants in this size range could cause severe pore blocking and significantly lower the membrane performance, which might be why the TMP rose rapidly with the inclined plate configuration used.

Figure 6 shows the morphological structures of a clean membrane and fouled membranes. Compared with the clean membrane (Figure 6(a)), the membranes in MC1, MC2 and MC3 were seriously fouled, indicated by compact cake layers formed on the used membrane surfaces, as shown in Figures 6(b)–6(d). Large cracks can be observed on the cake layer in MC1, as shown in Figure 6(b). These cracks may have been related to the effective looseness of the hollow fibers. As noted above, the lower end of the hollow fibers was not fixed. In MC1, there was no fetter around the bundle, and thus the hollow fibers were able to swing freely under the action of air scouring. Intense vibration of the hollow fibers exerted a force on the cake layer, and thus the cake layer was split up. Compared to dense cakes, these cracks provided easier access for fluid to penetrate the membrane pores, which was why the TMP rose relatively slowly when no auxiliary floc separation device was used in the membrane module (MC1). Research indicates that membrane performance could be enhanced when a degree of looseness of the hollow fibers was allowed in the gas-liquid two-phase flow (Wicaksana et al. 2006; Yeo et al. 2007). Figure 6(d) illustrates the presence of many small pores on the cake layer, which generally means good permeability. However, internal pore blocking
occurred in MC3 considering the rapid TMP development and high UMFI value. This conclusion is consistent with the inference in the previous analysis.

**Optimization of air scouring for the stacked inclined plate configuration**

As for the three membrane configurations discussed above, configurations similar to MC1 have been extensively studied (Cui et al. 2003; Yeo et al. 2006, 2007). Thus, only a limited discussion on MC1 is provided here. Liu et al. (2014) studied the performance of a cylindrical floc separation device. They investigated the membrane performance, floc properties and fouling potential under different bubbling conditions. The influence of air scouring was found to be multiple: bubbling-induced shear rate reinforced the vibration of the hollow fibers and slowed down the TMP development, however, bubble-enhanced mass transfer resulted in severe particle adsorption on the membrane surface and increased membrane resistance. Next, the performance of the newly made floc separation device MC3 is discussed in detail. The membrane performance of MC3 under different air scouring rates was studied and the effluent water quality was compared.

Five different gas flow rates were applied, and the corresponding effluent water qualities were measured. The results indicated that there is a fluctuation in the effluent water qualities under different gas flow rates, but none of the
cases exhibit a notable advantage against the others. There was a decrease in UV$_{254}$ in some cases, but the turbidity was high. The gas flow rate has only a slight effect on the effluent water quality in such a condition.

Figure 7 illustrates that the gas flow rate has a significant influence on TMP development. However, the influence of the gas flow rate was not unidirectional, which does not correspond to the finding of Tian et al. (2010). The TMP under 60 mL/min showed a minimum growth rate, indicating light membrane fouling. The fastest TMP growth occurred in the 15 mL/min bubbling case. The rapid membrane fouling may be due to the enhanced interaction between the flocs and hollow fibers caused by the bubbling disturbance. However, the surface shear stress induced by air scouring was weak under such a gas flow rate. The negative effect of bubbling dominated, and the net effect was that membrane fouling became worse. When a gas flow rate of 100 mL/min was applied, TMP growth also accelerated compared to the no-bubbling case, particularly in the last few hours of the filtration cycle. These results indicated that either an overly high or overly low gas flow rate could have detrimental effects on membrane performance.

The results based on the UMFI value were quite similar to those from TMP development. The UMFI in the 60 mL/min bubbling case was the lowest, with an average value of 6.02 m$^{-1}$. Combined with the TMP analysis, 60 mL/min is considered the optimal gas flow rate in terms of membrane fouling control. The decline of the TMP growth rate and UMFI value under a gas flow rate of 60 mL/min means that the positive effect of air scouring dominates. In the last few hours of the filtration cycle, pore blocking reached equilibrium, and the TMP became stable (shown in Figure 7(a)). The membrane performance deteriorated when 15, 35 and 100 mL/min of air scouring was applied, as indicated by the TMP and UMFI results. Liu et al. (2014) also found that air bubbling with a gas flow rate of no more than 40 mL/min accelerated membrane fouling, and they attributed this result to the high membrane resistance of the concentration polarization and adsorption. The high degree of membrane fouling in the 100 mL/min bubbling case might be related to the severe inner pore blocking by small flocs and colloids broken by intense gas sparging.

**CONCLUSIONS**

This paper proposed the concept of separating flocs in the UF membrane module and developed a novel floc separation device. The performance of three different membrane configurations was compared in the coagulation-UF and coagulation-air scouring-UF process, respectively. The main findings are as follows:

1. In the coagulation-UF process, there was no significant difference between the effluent water qualities of the three membrane configurations. The configuration without an auxiliary floc separation device (MC1) and the cylinder configuration (MC2) showed relatively low UMFI values and TMP growth rates.
2. In the coagulation-air scouring-UF process (with an air scouring rate of 35 mL/min), although the TMP and UMFI values were not considerably limited by the inclined plate configuration (MC3) used, there were
some advantages in terms of better effluent quality (lower UV$_{254}$) and good integrity of the flocs, which is favorable in membrane filtration processes. The configuration without an auxiliary floc separation device provided a good measure for hollow fiber movement, and the cake layer formed was porous. In addition, air scouring was effective in limiting TMP development in long-term operation.

3. When the inclined plate membrane configuration was used in the coagulation-air scouring-UF process, either an overly low or overly high gas flow rate could cause deterioration of the membrane performance, and the optimal gas flow rate was determined to be 60 mL/min.

4. The membrane module configurations have significant effects on the membrane performance in the gas-liquid two-phase flow membrane process by affecting the floc properties and hydraulic conditions. Further work should be performed to reveal the flow regimes and the interactions between the flocs, floc separation devices and hollow fibers in the hybrid process, and thus improving the membrane configuration will play a more significant role.

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REFERENCES


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