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Fouling behavior and system performance in membrane bioreactor introduced by granular media as a mechanical cleaning effect on membranes

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ABSTRACT

The use of granular polyethylene terephthalate (PET) media provides an effective way to control membrane fouling in aerobic membrane bioreactor (MBR) system under relatively low aeration rate in treatment of synthetic wastewater and real sewage. Without PET media, increasing set point permeate flux caused disproportionate increase in fouling rate. The fouling rate observed at higher flux could not be reduced by lowering the flux due to the formation of irreversible membrane fouling. At fixed aeration rate and permeate set point flux, the fouling rate was much lower with the flat sheet membrane than the hollow fiber membrane as it is combined with the PET media under 5% of packing ratio. The benefit of the granular PET media as suspended carriers to reduce membrane fouling was more pronounced with the flat sheet membrane than the hollow fiber membrane possibly due to better accessibilities and movements of the PET media within flow channels between membranes. However, the use of longer fiber improved beneficial effect of the PET media on fouling reductions than shorter one. It was thought that longer fiber enhanced fiber movement and this should provide synergistic effect on the use of PET media as mechanical cleaning way to control membrane fouling. During the operational period of MBR system, the addition of the rigid spherical PET media in the reactor did not deteriorate treatment efficiency, yielding more than about 97 and 75% in soluble Chemical oxygen demand (COD) and total nitrogen removal efficiencies, respectively.

Keywords: Membrane bioreactor; Membrane fouling; Granular media; Fiber length

1. Introduction

Membrane technology can offer the potential to increase the value of reclaimed wastewater significantly [1]. Recently, membrane technology becomes essential to meet regulatory water-quality standards in domestic wastewater treatments for reuse purposes. Unlike other reuse technologies, membranes can be specified to remove different classes of contaminants on the basis of membrane pore sizes, but they can also remove the contaminants on the basis of their chemical characteristics. Submerged membrane bioreactors (MBR) are the combined biological and physical process technology integrating suspended growth

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bioreactors with submerged membrane filtration [2]. There has been upsurge of interests in the MBR technology to improve the value of the reclaimed wastewater in domestic wastewater treatment processes. The MBR has several advantages over conventional activated sludge process since it can provide excellent effluent (permeate) qualities and smaller footprint, requiring lower hydraulic retention time (HRT) at relatively longer solid retention time.

However, membrane fouling caused by the deposition of the foulant materials on membrane surface and/or within membrane pores is an inevitable phenomena and major hurdle to be resolved. Although, the MBR technology has been proven successfully in wastewater reclamation process, membrane fouling can reduce membrane performance progressively, requiring periodic membrane cleaning and thus shortening membrane lifetime. Aeration has been widely adopted to induce both air bubbles and water movements around the bubbles along the membrane surface to provide hydraulic shear to reduce membrane fouling. However, the aeration is still energy intensive process in aerobic MBR system and thus may not be enough to remove sticky materials caused by microbial byproducts attached on membrane surface. It has been known that the aeration rate needed to control the membrane fouling should be about two or three times higher than that to sustain biodegradation in MBR technology [2-4].

In order to reduce aeration rate required while inducing better fouling control, additional cleaning strategies using granular materials for physical scouring actions on membrane surface can be considered. In many studies, it has been known that cake layer formed on membrane surface could play an important role in determining fouling resistance against water flows through the membranes in the MBR system. The use of air bubbling alone in the MBR system provides gas/liquid two phase flows along the membrane to mitigate membrane fouling. However, the fouling control varies depending upon various characteristics of air bubbles such as aeration rate and bubble shapes [5]. Many studies have demonstrated that membrane flux tended to increase with aeration rate, but up to plateau value, which implies that no further reduction in membrane fouling is achieved beyond the optimal aeration rate [1,2,5,6].

This study deals with the use of the plastic spherical media combined with aeration to ensure fouling control and system performance in MBR system applied for wastewater reclamation purposes. The movement of the granular media combined with aeration can be expected to provide synergistic effects on the control of membrane fouling. Continuous movements and physical contacts of the media with membrane surface can limit concentration polarization layer or cake layer formation on membrane surface at relatively lower aeration rate required. Interests in using suspended media to ensure long membrane lifetime at enhanced membrane flux are growing, but the use of the media as a mechanical cleaning tool for reducing membrane fouling in MBR system needs more attentions [7,8]. Better understanding the roles of the granular media on fouling control and permeability qualities is critical to improve design and operation of the media-based MBR system and optimize system performance and reduce costs further. The objective of this study is to investigate the effect of spherical media consisting of polyethylene terephthalate (PET) on membrane fouling and permeate qualities in aerobic MBR system treating low-strength wastewaters. Both short-term and long-term operations were conducted using laboratory-scale MBR system with the addition of the PET media combined with aeration at various operational conditions to observe the behavior of membrane fouling and system performances. Comparisons on fouling control observed under the suspension of the PET media between the hollow fiber membrane and flat sheet membrane modules were also made in the MBR system.

2. Methods

2.1. Experimental setup

Fig. 1 is a schematic diagram of the laboratory-scale MBR system used in this study. The MBR system consisted of anoxic and aerobic (AO) tanks for denitrification and nitrification process, respectively. Total effective volume of the AO MBR reactor was 10 L. Membranes consisting of polyethylene (PE) having 0.4 µm pore size were submerged directly into aerobic bioreactor to separate mixed liquor suspended solids (MLSS). The open sections of the membranes were connected by tubing to the peristaltic pump (Masterflex, Model No. YZ1515X-D) to drive the permeate flow from outside to inside of the membrane. Suction pressures at the open sections of the membrane were monitored by pressure gauge periodically to observe membrane fouling. Internal recycle line was placed between anoxic and aerobic reactor at 2 Q of recycle ratio. Diffusers were installed at the bottom of the aerobic reactor to provide air bubbles to control membrane fouling and oxygen supply. Air flow rates tested in this study ranged from 2.5 to 10 L/min. The oxidation/reduction potential, dissolved oxygen concentration, solution pH, and temperature in reactors were monitored. All data were recorded by data acquisition



Fig. 1. Schematic diagram of experimental setup.

system. Temperature in aerobic reactor was maintained at about 25°C using temperature controller submerged in the reactor. Synthetic feed solution was delivered to the anoxic tank with a peristaltic pump (Masterflex, Model No. YZ1515X-D) at a flow rate controlled to maintain a constant water level in the reactor automatically. Total HRTs were 8, 12, and 24 h depending upon the permeate flux of the membranes applied. Permeate flux of hollow fiber membrane was 12, 8, and $4 \text{ L/m}^2/\text{h}$ which correspond to 8, 12, and 24 h of total HRT, respectively. With flat sheet membrane module, peristaltic pump was connected at the open ends of the membranes to pump at 41, 27, and $14 \text{ L/m}^2/\text{h}$ for the same HRTs. The feed suspension for MBR system consisted of glucose at equal chemical oxygen demand (COD) concentrations of 200-300 mg/L. The NH₄Cl and Na₂PO₄ were added as source of nitrogen and phosphorous. Compositions of feed solution tested for this study are summarized in Table 1.

Table 1 Compositions of synthetic wastewater

Composition	Concentration (mg/L)
Glucose	187.5
NH ₄ Cl	136.42
Na ₂ HPO ₄	18
NaH ₂ PO ₄	16
FeSO ₄	0.25
MgSO ₄ ·7H ₂ O	23
NaCl	12

In this study, the effluent (permeate) produced by the MBR system was further treated by reverse osmosis (RO) membrane intermittently. Cross-flow filtration membrane unit (Osmonics, GE) was operated under the constant pressure of 10 bar. Both permeate and concentrate stream produced from the cross-flow membrane unit was recycled back into the feed tank to maintain water level in it. Temperature controller was installed in the feed tank to prevent the increase in water temperature caused by running high pressure pump to range from 15 to 20°C in feed water.

2.2. Membranes and granular media

In this study, both hollow fiber membrane and flat sheet membrane module were tested in the MBR system. PE hollow fiber membranes with nominal pore size of 0.4 µm and total membrane surface area of 0.1 m² were tested. Flat sheet membranes composed of polyvinyle chloride were also tested for observing effect of granular media on membrane fouling. A total membrane surface of the flat sheet membranes was 0.04 m². Granular materials consisting of PET were applied as suspended media in aerobic MBR to control membrane fouling as shown in Fig. 1. The diameter and specific gravity of the PET media were 3 mm and 1.3, respectively. About 5 and 10% of the packing ratio of total volume of aerobic MBR reactor were applied with aeration provided at the bottom of reactor. The spherical PET media were introduced into the aerobic reactor and flowed upward with air bubbles along the membrane surface and returned downward outside the membrane module.

2.3. Analytical methods

COD, total, and volatile suspended solids (TSS and VSS) were measured according to Standard Method. For soluble chemical oxygen demand (SCOD) analyses, samples were perfiltered using 1.2 mm glass fiber filter. The same glass fiber filters were used for TSS and VSS measurements. Total nitrogen (TN) and phosphorous in the samples were measured using spectrophotometer (DR2800, HACH).

3. Results and discussion

3.1. Set point flux and membrane fouling

The MBR system was seeded with the activated sludge taken from a local domestic wastewater treatment facility and fed with synthetic wastewater continuously. Initial permeate flux for the hollow fiber membrane submerged in the reactor was set as 4 L/m^2 /h which corresponded to 24 h of total HRT. The length of the hollow fiber membrane tested was 0.1 m. Variation of TMP with time obtained from the operation of the aerobic MBR system is presented in Fig. 2. During the acclimation period of the MBR system, the PET media were not added into the reactor in which only aeration was supplied at 5 L/min from the bottom of the reactor. At a set point permeate flux of 4 L/m^2 /h, the suction pressure was maintained below 0.1 bar during 15 d operation. Increasing set point flux



Fig. 2. Variation of TMP with time in hollow fiber membranes at different set point fluxes (aeration rate: 5 L/min, no addition of PET media).

to $6 L/m^2/h$ increased suction pressure to 0.25 bar within a week, but lowering back to $4 L/m^2/h$ did not recover original value of the suction pressure observed at the same flux (0.1 bar) probably due to the formation of irreversible fouling. During 30 d operation, the MLSS concentration was maintained as about 3,000 mg/L. At 35 d, the permeate flux then increased from 4 to 8 $L/m^2/h$. At 8 $L/m^2/h$, the TMP increased from 0.15 to 0.3 bar within 7 d operation. In order to maintain 8 h of total HRT, the permeate flux increased further to $10 \text{ L/m}^2/\text{h}$ after which sudden jump in TMP to 0.45 bar was observed within a day. Subsequently, membranes were taken from the reactor to perform recovery cleaning at 42 d. The MLSS concentration was about 5,500 mg/L as first chemical cleaning with used membranes was attempted.

At 45 d of operation, the MBR system was operated subsequently at 8 h of total HRT and at that time was increased MLSS concentration to about 5,500 mg/L. Both membrane relaxation and backwashing were not applied during the operational period of the MBR system. Results in TMP changes with time after 45 d are presented in Fig. 3. In this test, both hollow fiber membrane and flat sheet membranes were compared without PET media under the aeration only. Without PET media, the TMP increased rapidly to 0.4 bar within 7 d operation and this required weekly based chemical cleaning. At 72 d, hollow fiber membrane was replaced with the flat sheet membrane in the same MBR reactor, and then operated the reactor subsequently. With flat sheet membrane, the TMP increased



Fig. 3. Long-term observations of TMP change with flat sheet and hollow fiber membrane module (airflow rate: 5 L/min, HRT: 8 h, packing ratio of PET media: 10%). Note: A: hollow fiber membrane without media, B: flat sheet membrane without media, C: flat sheet membrane with media and D: hollow fiber membrane with media.

to 0.3 bar at 93 d of operation. Fouling rate with flatsheet membrane was somewhat slower than that observed with hollow fiber membrane, but the aeration rate itself without PET media was not very effective to maintain TMP value below 0.3 bar.

At 93 d, the PET media were added with 10% of packing ratio into aerobic MBR reactor at which flat sheet membranes are submerged under 5 L/min of aeration rate. Result shows clearly that the addition of PET media as suspended carriers into the MBR reactor reduces membrane fouling significantly. The TMP increased rapidly to about 0.1 bar within a day, but no further increase in TMP with time was observed during the operational period of the MBR reactor. At 123 d, the flat sheet membrane was replaced with the hollow fiber membrane to further investigate the effect of the PET media on membrane fouling. As shown in Fig. 3, the beneficial effect of the PET media on fouling reduction was very limited with the hollow fiber membrane as compared to the flat sheet membrane. For both flat sheet and hollow fiber membranes, the foulants causing initial membrane fouling were not removed effectively by suspending PET media along the surfaces of the membranes, suggesting the PET movement may not be enough to remove initial fouling mostly caused by pore fouling. The fact that PET media limit control for the initial fouling may be related to the types of foulant materials. Pore fouling is resulted from the small particles and/or colloidal materials that approach into the membrane pores and it can increase membrane resistance [9]. The initial fouling can be caused mainly by the pore blockage and/or pore adsorption within membrane pores against which the contact of the PET media along membrane surface should not be very effective to remove these small colloidal material away from membranes. Nevertheless, sudden jump in TMP was not observed with the flat sheet membrane during the operational period of the MBR system. This indicates that the build-up of fouling layer on membrane surface can be prevented effectively by the physical scouring actions induced by the movement of the rigid PET media on the surface of the membrane.

3.2. Treatment efficiency

Results in SCOD and TN concentrations in feed solution and membrane permeate during operational period with MBR system are presented in Figs. 4 and 5, respectively. At start up of operation, the MBR system was fed with synthetic wastewater consisting of 200 mg/L of SCOD. After 45 d operation, the SCOD in feed solution increased to 300 mg/L to increase organic loading rate to 0.01 kg/d. During operational period, the average SCOD removal in MBR system



Fig. 4. SCOD concentrations with time in feed solution and permeate from MBR.



Fig. 5. TN concentrations with time in feed solution and permeate from MBR.

was 97.7%, yielding average SCOD concentration in membrane permeate was about 6.9 ± 3.3 mg/L. Replacement of hollow fiber membrane to flat sheet membrane at 45 d operation did not change SCOD removal efficiency significantly. Concentration of suspended solids in membrane permeate was near zero due to the small pore sizes of both membranes (0.4 µm) tested. After addition of PET media into aerobic MBR reactor at 93 d operation, permeate qualities were measured periodically. Since the PET media were added into the aerobic MBR reactor, the MLSS concentration was maintained as about 8,000 mg/L. After 60 d operation, TN removal was maintained as about 73.7%, yielding $10.6 \pm 2.2 \text{ mg/L}$ in membrane permeate. The MBR permeate was further treated by RO membrane and the average TN and total phosphorus concentrations in RO permeate were 1.2 and 0.02 mg/L, respectively. This indicates that the effluent qualities from the integrated MBR-RO system be enough to meet the water reuse standards sufficiently. During overall operational period, the addition of the PET media into the MBR system did not provide any negative effects on the permeate qualities from the reactor.

3.3. Short-term tests on the effect of PET media on fouling with flat sheet membrane

In order to further investigate the effect of PET media on membrane fouling, short-term experiments were performed with the flat sheet membrane applied with the real mixed liquor suspension taken from the pilot-scale MBR system operated at local wastewater treatment facility. For this test, the membrane reactor was operated as a total recycle mode where permeate produced by the membrane was recycled back into the reactor to maintain constant volume of feed water. During these experiments, the MLSS concentration taken from the pilot-scale MBR was about 11,000 mg/L and then diluted to 5,500 mg/L prior to performing laboratory short-term experiments. The packing ratio of the PET media and set point permeate flux were 10% and 25 L/m²/h, respectively. Results are presented in Fig. 6. Under the aeration itself without PET media, the TMP approached to 0.12 bar at 10 h of filtration. However, the addition of the PET media under the same aeration rate of 5 L/min maintained TMP value below 0.08 bar during the same filtration time, suggesting that fouling rate be reduced by the movement of the PET media along the surface of flat sheet membrane effectively. This was also confirmed by the results observed in the long-term operation of the laboratory-scale MBR system (see Fig. 3).

Subsequently, the effect of the PET media on fouling control was further investigated using increased MLSS concentration of 11,000 mg/L as feed suspension. In this test, experiments were performed first without PET media at two different aeration rates of 5 and 10 L/min. As shown in Fig. 7, under the aeration only without PET media, was initial fouling rate very rapid, but after 2 h filtration, fouling rate approached pseudo-steadystate condition at both aeration rates (5 and 10 L/min). Under the same aeration rate of 5 L/min, the addition of the PET media reduced fouling rate almost by two times as compared to the results observed without the media under the same aeration rate. The TMP value under the movement of PET media at 5 L/min of aeration rate was marginally lower than that observed at higher aeration rate of 10 L/min without the PET media. This suggests that the use of PET media can reduce energy cost required for aeration to reduce membrane fouling significantly. Because specific gravity of the PET media tested was about 1.3, there may be an existence of critical aeration rate beyond which the PET media can be movable along the surface of membrane without settling of them. However, due to the limit of the air flow range which can be measured by gas flow meter and bubbling produced by laboratory-scale air diffusers used, critical aeration rate could not be observed in this study.

The need to reduce membrane fouling is one of the major costs including capital and operational costs in submerged MBR system [3]. The aeration is widely applied for reducing membrane fouling as air bubbles can limit membrane fouling outside submerged membranes by inducing surface shear near to the fibers, lateral liquid flow across vertical fibers and later fiber movement due to fiber flexibility [10]. Nevertheless, supplying air bubbles require about half of total energy to operate MBR system. The use of granular materials combined with aeration can have an important beneficial effect on scouring membrane surface mechanically as observed in this study. Although, understanding dominant fouling reduction mechanisms in MBR combined with granular media needs more attentions, the addition of PET media tested in this study should improve additional shear conditions on the membrane surface, thereby preventing the build-up of the fouling layer on membrane surface more effectively when compared to the fouling control caused by the aeration itself.

3.4. Combined effect of PET media and fiber length on fouling control

The effect of PET media on TMP behavior with time for two different lengths of the hollow fiber membranes was investigated. The results for 0.1 m fiber length are presented in Fig. 8. Unlike flat sheet membrane (Fig. 7), instantaneous TMP behavior at 25 L/m²/h was very similar regardless of the addition of PET media, suggesting there is no beneficial effect of the PET media on fouling reduction in hollow fiber membranes. In the absence of PET media, aeration rate was increased from 5 to 10 L/min to investigate fouling rate under the same permeate flux of 25 L/m²/h. Without PET media, higher aeration rate did not cause further reduction in membrane fouling. This result may be caused by short bundle of the fibers by which contact of air bubbles to membrane surfaces can be limited or convective flux of foulants toward membrane surface is still lower than their back transport away from the membrane due to aeration. Although permeate flux was reduced to $12.5 \text{ L/m}^2/\text{h}$, there was no difference in transient behavior of TMP between with and without PET media in membrane reactor (data are not shown).

The fact that either aeration or PET media could not control membrane fouling in the bundle of hollow fiber membranes effectively may be related mainly to the manner in which the hollow fiber membranes are displaced. In aerobic MBR system, fiber movement has been reported as important factor to control membrane fouling and this effectiveness is more pronounced with longer fiber or smaller inside diameter of hollow fiber membrane [10]. In order to further investigate the effect of fiber length on membrane fouling, membrane module consisting of the longer fiber length which is 0.3 m was applied for the treatment of the MLSS having 11,000 mg/L. As shown in Fig. 9, the beneficial effect of the PET media on the reduction of membrane fouling is very significant for longer fiber. Under the movement of the PET media at 5% of packing ratio, the TMP value was maintained below 0.04 bar during the whole operational period of the MBR system using 0.3 m fiber length under the 5 L/min of aeration rate. Furthermore, at the same packing ratio of PET media (5%), lowering the aeration rate to 2.5 L/min allowed TMP value only near 0.05 bar at 24 h operation. Although packing ratio was reduced to 2.5%, lowering aeration rate did not increase TMP values significantly.

These results indicate that the use of the PET media in aerobic MBR system can reduce membrane fouling and energy cost required for the aeration to control membrane fouling. The results presented in Fig. 9 also suggest that the effect of PET media on fouling control in submerged MBR system can be dependent upon the fiber length significantly. Since the PET media need to be suspended along the membrane with help of aeration, beneficial effect of cleaning actions by PET media is related directly to the increasing bubble-induced benefit to control membrane fouling in MBR system.

The beneficial effect of the PET media on membrane scouring can vary depending upon the types of the membrane modules such as fiber length. Improvements in the design of membrane module and operation of it will be thus led by better understanding of these effects. Importantly, accessibility and contact of the PET media on the whole areas of the membranes should be the main factor to expect synergistic effect of PET media on fouling control. As shown in Figs. 6 and 7, the flat sheet membrane causes more positive effect on fouling control than the hollow fiber membrane under the movement of PET media. Flow channels between flat sheet membranes may be defined better than the bundle of hollow fiber membranes. This allows better approach of the PET media along membrane surface. Therefore, optimizing membrane module design is a prerequisite to maximize the



Fig. 6. Effect of PET media on TMP change with time for flat sheet membrane (set-point flux: 25 L/m^2 /h, aeration rate: 5 L/min, packing ratio of the PET media: 5%, MLSS: 5,500 mg/L).



Fig. 7. Effect of PET media on TMP change with time for flat sheet membrane (set-point flux: $25 \text{ L/m}^2/\text{h}$, packing ratio of the PET media: 5%, MLSS: 11,000 mg/L).

effectiveness of the granular materials as they are applied for the mechanical cleaning tool for reducing membrane fouling in MBR system.

Since granular PET media are suspended under the aeration provided at the bottom of the reactor, the effect of mechanical cleaning of the PET material on fouling control should be closely related to the bubble-induced behavior. Based upon the results observed in this study, the use of longer hollow fiber can be more beneficial than the shorter one in term of fouling control as the PET media is introduced in MBR. Wicaksana et al. suggested that fiber movement be increased with longer fibers due to higher chances of fiber flexibility [10]. In typical aerobic MBR system, while the amplitude of the movement for longer fibers is expected to be higher, fouling mitigation effect can



Fig. 8. Effect of PET media on TMP change with time for hollow fiber membrane (set-point flux: $25 \text{ L/m}^2/\text{h}$, packing ratio of the PET media: 5%, MLSS: 11,000 mg/L, fiber length: 0.1 m).



Fig. 9. Effect of PET media on TMP change with time for hollow fiber membrane (set point flux: $12.5 \text{ L/m}^2/\text{h}$, MLSS: 11,000 mg/L, fiber length: 0.3 m).

also be generally lower with longer fiber length due to larger lumen pressure losses [10,11]. Nevertheless, the use of PET media as mechanical cleaning tool may be able to mitigate local fouling caused by the longitudinal pressure drop along the fiber length as well. As shown in Fig. 10, the membrane fouling after 24 h filtration was considerable near two open ends of fibers where suction pressure is expected to be highest. However, the addition of PET media cleaned membrane fouling along the fiber length effectively. Lowering packing density of hollow fibers within membrane module can be another possible way to improve media effect on fouling control. However, packing densities based upon the potting areas for 0.3 and 0.1 m fiber modules tested in this study were 0.12 $0.09 \text{ m}^2/\text{m}^2$, respectively. and Therefore, fiber



Fouling under aeration only

Fouling under aeration and PET media

Fig. 10. Images of fouling along fiber length after 24 h filtration experiment (fiber length: 0.3 m, set-point flux: $12.5 \text{ L/m}^2/\text{h}$, packing density of PET media: 5%, aeration rate: 5 L/min, MLSS: 11,000 mg/L).

flexibility appears to be more important than packing density to improve the effectiveness of PET media on fouling control. From our long-term operation with the MBR system combined with granular material, the addition of the PET media did not deteriorate qualities of membrane permeates, but selection of media material should also be important to safe microbial activities. Although the use of granular media is promising in terms of fouling control and energy savings in MBR system, optimizing reactor design, membrane module, and membrane materials needs to be studied as future works more extensively.

4. Conclusions

This study points to the effect of PET media as mechanical cleaning tool on the membrane fouling and airflow rate required from experimental observations from the laboratory tests using submerged MBR systems treating low-strength wastewaters. The addition of the PET media greatly reduced the rate of TMP increase with time for flat sheet membrane, although the added benefit was diminished as short hollow fiber membrane was applied. In comparison to short hollow fiber, the use of longer fiber showed better fouling control by introducing PET media. It was found that aeration rate required to reduce membrane fouling could be reduced significantly by adding the PET media due to enhanced scouring actions along membrane surfaces. The addition of the PET media in aerobic MBR did not deteriorate effluent qualities from MBR system. The importance of the fiber length was also shown by attempts to compare fouling rate with respect to two different fiber lengths. Lower membrane fouling with longer fiber length under the movement of the PET media may originate from higher flexibility and displacement of the hollow fiber.

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