Influence of different biofilm carriers on the operation and membrane fouling of submerged membrane bioreactors

Tokihiro Kurita, Takuma Mogi, Katsuki Kimura*

Division of Environmental Engineering, Hokkaido University, N13W8, Kita-ku, Sapporo
060-8628, Japan

*Corresponding author

Tel: +81-(0)11-706-6271, E-mail address: kkatsu@eng.hokudai.ac.jp

Abstract

It has been reported that the combination process of biofilm and submerged membrane bioreactors (MBR) can mitigate membrane fouling. In this study, the influence of using three different biofilm carriers (fixed rope carrier, moving granular carrier and moving sponge carrier) on the operation and membrane fouling of MBRs was investigated. Using rope or sponge carriers with MBR processes improved the removal efficiency of nitrogen via the creation of an anoxic part inside the carriers. However, membrane fouling became very severe when rope carriers were used. Using granular or sponge carriers effectively mitigated membrane fouling because they mechanically cleaned the membrane surface. The cake/gel layer on the membrane surface was perfectly removed by granular or sponge carriers. However, the deterioration of the filterability of the mixed liquor suspensions was significant with granular carriers, leading to the evolution of physically irreversible fouling, whereas such deterioration was insignificant with the sponge carrier. The characteristics of the soluble microbial products (SMP) significantly varied depending on the types of carriers used, which could influence the filterability of mixed liquor suspensions and the development of irreversible membrane fouling.

Keywords

Membrane bioreactor (MBR), Biofilm carrier, Membrane fouling, Filterability of mixed liquor suspensions

1. Introduction

Introducing granular or sponge materials into submerged membrane bioreactors (MBRs) can mitigate membrane fouling and bring about a stable operation because of their mechanical cleaning effect on the membrane surface [1,2]. Some researchers have demonstrated that the

introduction of granules or sponge materials into MBRs effectively removes the cake/gel layer from the membrane surface [3–6]. Additionally, a previous study indicated that the introduction of granules could reduce 50% of the aeration rate in a pilot-scale MBR [7]. Thus, the introduction of granules or sponges into submerged MBRs has the potential to significantly reduce the operation cost.

Some of the materials examined in previous studies were originally developed as biofilm carriers in wastewater treatment. Therefore, when introducing those biomass carriers into MBRs, a certain amount of biomass should be present as biofilm, which may affect the process performances of MBRs. The possession of biofilms in MBRs improved the removal efficiency of nutrients compared to conventional MBRs [1,8,9]. Various types of biofilm carrier configurations are commercially available, including fixed rope or mesh material hung in the reactor [10,11], a gear or cylindrical ring shape of the plastic material suspended in the reactor [4,9,12,13], and granules/sponge suspended in the reactor. In some studies, the fluidizing space for the carriers was separated from the membrane tank to prevent damage to the membrane surface [9,12–15]. It has been reported that the combination of biofilm and the MBR process mitigates membrane fouling by improving the filterability of mixed liquor suspensions [1,4,14–16]. In contrast, some researchers have noted that this combination process induced membrane fouling because of the promotion of the accumulation of cake on the membrane surface [17,18]. These contradicting results suggest that the configuration and/or type of biofilm carriers (moving or fixed in the reactor) used in the combination process influence the characteristics of membrane fouling. To choose an optimum biofilm carrier for the mitigation of membrane fouling in the combination process, the influence of the properties of the biofilm carrier on the characteristics of membrane fouling need to be investigated. However, a comparison among different types of biofilm carriers in terms of the control of the fouling in MBRs has rarely been conducted.

The objective of this study was to investigate the influence of different biofilm carriers on the operation and membrane fouling in MBRs. In this study, a fixed rope carrier, a granular carrier and a sponge carrier were examined. In the cases of granular or sponge carriers, the carriers freely moved in the reactor and made contact with the membrane surface. In contrast, rope carriers were immobilized in the reactor; there was no contact between the carriers and the membrane surface. Bench-scale MBRs with the above-mentioned biofilm carriers were operated continuously side by side with synthetic wastewater. The filtration resistance and components of mixed liquor suspensions in each MBR were investigated. In addition, the characteristics of membrane fouling were analysed and compared from various aspects.

2. Materials and methods

2.1 Operation of bench-scale MBRs

In this study, four identical bench-scale MBRs were operated in parallel. The effective volume of the bench-scale MBRs was 7.5 L. Three MBRs were operated with different biofilm carriers: a rope carrier (denoted as MBR-A hereafter), a granular carrier (MBR-B), and a sponge carrier (MBR-C). The remainder was operated without a carrier (Control-MBR). The rope carrier (RingLace, Kajima Corp., Japan) consisted of polyvinylidene fluoride (PVDF) fibre strings that were loosely woven into the form of a rope. The outer diameter of this carrier was 25 mm. The rope carrier was cut into a length of 10–15 cm and hung in the MBR vertically. The total length of rope carriers in MBR-A was 2.5 m, in accordance with the recommendation of the manufacturer, 0.33 m/L of tank volume. Degradation and dissipation of the rope carrier were not observed during the operation. The granular carrier (BCN, Nisshinbo Chemical Inc., Japan) had a cylindrical shape and was used by the authors in previous research [5]. The size (both diameter and height) and specific gravity were approximately 4 mm and 1.01, respectively. The main component of the granular carrier was polyethylene glycol. The sponge carrier (Achilles Corp., Japan) used in this study was made from polyether-urethane foam and had a cubic shape. The density of the sponge carrier was 35 kg/m³ with 46 cells per 25 mm. The dimensions of the cubes were 4 × 4 × 4 mm. The apparent volume of the granular and sponge carriers introduced into the MBRs was 5% of the reactor volume, which was determined based on the authors' previous research [7]. The rope carrier and sponge carrier used in this study are used in integrated biofilm activated sludge (IFAS) and moving bed biofilm reactor (MBBR) applications [20]. The rope carriers were fixed in the reactor to prevent contact with the membrane surface. In the cases where granular or sponge carriers were used, the carriers moved freely in the reactor and made contact with the membrane surface.

Each MBR was equipped with 0.06 m² flat-sheet microfiltration membranes (Toray, Japan). The membrane was made from PVDF and had a nominal pore size of 0.1 µm. Aeration was performed continuously (aeration rate: 17 L/min) beneath the membrane modules. Intermittent operation (12 min operation was coupled with a 1-min pause) was carried out in this study. The membrane flux was set at 18.8 LMH. The resulting hydraulic retention time (HRT) was 7.2 h. A fixed amount of mixed liquor suspension (380 ml) was wasted daily from each MBR. As a result, the solid retention time (SRT) in Control-MBR was 20 days.

Properties of the membrane used in this study and operational parameters of the MBRs are summarised in Table 1. The temperature in the MBRs was maintained at 20 °C using a water bath. When membrane fouling became significant (trans-membrane pressure (TMP) > 40 kPa), the membrane modules were removed from the reactor and the membrane surface was manually wiped with a sponge. The bench-scale MBRs operated in this study were inoculated with sludge collected from a pilot-scale MBR installed at a full-scale municipal wastewater treatment facility (Soseigawa Wastewater Treatment Center, Sapporo, Japan). The synthetic wastewater used in this study was prepared by following the Organization for Economic Cooperation and Development (OECD) guidelines 302A and 303A [21]. The major carbon sources of this synthetic wastewater were peptone and a meat extract. Biomass was acclimatized to the synthetic wastewater for 40 days. One month after the acclimatization started, the mobility of the sponge carriers was significantly lowered because of the excess growth of biofilm. Therefore, an extra aerator was placed in the bottom of MBR-C and was intermittently used (20 s aeration with a 5-min pause) to facilitate the movement of sponge carriers.

Table 1 Properties of the membrane used in this study and operational conditions of the MBRs.

Membrane configuration	Flat sheet
Membrane pore size	0.1 µm
Membrane material	PVDF
Membrane surface are	0.06 m ²
Flux	18.8 LMH
Aeration rate	17 L/min
HRT	7.2 hours
SRT	20 days

2.2 Assessment of the filterability of the mixed liquor suspensions

At the termination of the operation, the filterability of the mixed liquor suspensions was assessed by a series of batch filtration experiments using a dead-end stirred cell system (Advantec Tokyo, Japan). The membrane used in these experiments was identical to that used in the bench-scale MBRs. The effective membrane area in the stirred cell was 37.4 cm². In each filtration, virgin membranes were used. The stirred cell was filled with 300 ml of mixed liquor suspensions, and compressed N₂ gas was used to filtrate the suspensions. The filtration

pressure was fixed at 10 kPa, and the stirring speed in the cell was set at 300 rpm. The membrane flux was measured using an electronic balance. The membrane filtration resistance was determined according to Darcy's law:

$$R = \Delta P / (J \cdot \mu)$$

where R is the membrane filtration resistance (m^{-1}), J is the membrane permeate flux ($\text{m}^3/\text{m}^2/\text{s}$), ΔP is the TMP (Pa), and μ is the viscosity of the permeate (Pa·s). In a series of batch filtration experiments, the membrane permeate flux reached a plateau when the filtration time exceeded approximately 20 min. Subsequently, 5 min of additional filtration was conducted, and the membrane filtration resistance (i.e., the filtration resistance of the mixed liquor suspensions) was calculated based on the average membrane flux during the additional filtration time.

2.3 Analytical methods

The concentrations of the total organic carbon (TOC) and dissolved organic carbon (DOC) were determined using a TOC analyser (TOC-VCSH, Shimadzu, Japan), which could also be used to determine the concentration of total nitrogen (T-N). Analysis of the DOC, dissolved polysaccharides and dissolved proteins in mixed liquor suspension samples was performed after the suspensions had been centrifuged ($2100 \times g$, 5 min) and filtered with a $0.45\text{-}\mu\text{m}$ mix cellulose ester (MCE) filter (Advantec Tokyo, Japan). The phenol-sulfuric acid method [24] and the Lowry method [23] were used to determine the concentration of polysaccharides and proteins, respectively. Glucose and bovine serum albumin (BSA) were used as standards for the measurements of polysaccharides and proteins, respectively. Analysis of the monosaccharide composition of dissolved polysaccharides was conducted using a Dionex DX 500 HPLC equipped with a pulsed amperometric detector (Dionex ED40) following the method described by Croué et al. [24]. The amount of biomass growth on the rope carriers was measured following the method described by Yang et al. [25]. The amount of biomass attached on the granular and sponge carriers was measured following the method described by Khan et al. [8].

In this study, four bench-scale MBRs were operated for 30 days. At the termination of the operation, the fouled membranes were collected from the reactors and analysed. The filtration resistances of the fouled membranes were measured using the same system as used in the filterability test described above (effective membrane area: 3.5 cm^2). In the measurements of the filtration resistances, distilled water was filtrated under a constant pressure (30 kPa), and the effectiveness of the different cleaning methods was investigated. Additionally, the surfaces

of the fouled membranes were observed using scanning electron microscopy (SEM) (S-4000, Hitachi, Japan). The membranes were cut into 1 cm² squares, dipped in 2% glutaraldehyde in 0.1 M phosphate buffer, and then transferred to 1% osmium tetroxide with 0.1 M phosphate buffer to fix the structure of the fouling layer. The fixed samples were dehydrated in ethanol and subsequently immersed in isoamyl acetate solution. The samples were then dried in a critical point drying apparatus (HCP-1, Hitachi, Japan) to avoid the deformation of the foulant structure. Finally, the samples were coated with ultrathin platinum to avoid the accumulation of a static electric charge during the observation.

3. Results and discussion

3.1 Quality of treated water

The mixed liquor suspended solids (MLSS) and attached biomass concentrations in each MBR are shown in Table 2. The MLSS concentration in the Control-MBR was lower than the others. In MBR-A, the amount of attached biomass was the largest, whereas the granular carriers had the least amount of attached biomass. Although the apparent volume of the granular and sponge carriers was the same (5% of the reactor volume), the amounts of attached biomass were significantly different in the two reactors.

Table 2 Concentrations of MLSS and attached biomass in the four MBRs.

(mg/L)	Control-MBR	MBR-A	MBR-B	MBR-C
MLSS	4000 ± 760	5100 ± 860	5400 ± 460	4800 ± 170
Attached biomass		780	50	240

The average concentrations of the TOC and T-N in the synthetic wastewater and treated water during the operation are summarized in Table 3. The TOC removal exceeded 94% in the four MBRs. It should be noted that MBRs operated with the carriers exhibited higher TOC removal. The highest TOC removal was observed with MBR-C. Regarding the T-N removal rate, MBR-A and MBR-C exhibited better performances than did the other two. In this study, the four MBRs were fully aerated and were not combined with denitrification measures (e.g., installing an anoxic tank or intermittent aeration). The rope and sponge carrier held larger amounts of attached biomass than did the granular carrier (Table 2). Liu et al. [9] and Khan et al. [8] reported that the deeper zone of the biofilm carrier used with MBRs created an anoxic

zone in which denitrification was facilitated. It is thought that an anoxic zone was also created in MBR-A and MBR-C, leading to the high T-N removal rates.

Table 3 Performance of the MBRs.

	Control-MBR	MBR-A	MBR-B	MBR-C
TOC influent (mg/L)	160.4 ± 19.9	160.4 ± 19.9	160.4 ± 19.9	160.4 ± 19.9
TOC permeate (mg/L)	8.5 ± 3.6	7.3 ± 2.1	7.5 ± 1.7	6.1 ± 1.0
TOC removal rate	94.7%	95.4%	95.3%	96.2%
T-N influent (mg/L)	71.1 ± 9.1	71.1 ± 9.1	71.1 ± 9.1	71.1 ± 9.1
T-N permeate (mg/L)	66.7 ± 8.1	58.6 ± 10.5	67.3 ± 3.7	57.1 ± 6.1
T-N removal rate	6.2%	17.5%	5.3%	19.7%

3.2 Influence of different carriers on the sludge characteristics

Table 4 summarizes the average concentrations of dissolved organic matter in each MBR. The dissolved organic matter in the MBRs was mostly composed of soluble microbial products (SMP) [26]. Although a sufficient acclimatization period was provided prior to the continuous operation, the concentrations of the dissolved organic matter rapidly increased after 20 days in the Control-MBR. In contrast, the concentrations of dissolved organic matter were stable in the other MBRs operated with biofilm carriers. The use of biofilm carriers might function to stabilize the sludge characteristics in MBRs. In the cases of MBR-A and MBR-C, which had larger amounts of attached biomass than MBR-B, the concentrations of the dissolved organic matter (especially the concentration of dissolved polysaccharides) in the reactors were lower than the other two MBRs. It is known that the production of SMP is lowered under anaerobic conditions compared to aerobic conditions [27]. The bottom section of the biofilm attached on the rope and sponge carriers became anaerobic and might contribute to the low concentrations of the dissolved organic matter in the two reactors. Liu et al. [9] reported that the dissolved organic matter was adsorbed on the developed biofilm in an MBR. This could also contribute to the low concentrations of dissolved organic matter in MBR-A and MBR-C. In contrast, MBR-B exhibited higher concentrations of dissolved organic matter in the reactor than did MBR-A and MBR-C. As shown in Table 2, the amount of attached biomass was small in the case of MBR-B. Therefore, the reduction in the production of SMP and/or adsorption of SMP onto the biofilm could not be significant in the case of MBR-B. Additionally, Huang et al. [3] reported that the granular carriers broke sludge flocs and that some extracellular polymeric substances (EPS) were released to the supernatant

of the mixed liquor suspensions, leading to the elevation of the concentration of dissolved organic matter in the reactor.

Table 4 Average concentrations of dissolved organic matter in the four MBRs.

	Control-MBR		MBR-A	MBR-B	MBR-C
	Before 20 d operation	After 20 d operation			
Polysaccharide (mg/L)	64.5 ± 14.2	72.7 ± 12.5	48.9 ± 8.1	67.4 ± 6.7	37.7 ± 5.8
Protein (mg/L)	14.5 ± 2.0	31.8 ± 7.3	14.7 ± 2.2	18.9 ± 2.6	11.8 ± 2.2

3.3 Membrane fouling behaviour during continuous operation

The time course changes of TMP in each MBR are shown in Fig. 1. Physical cleaning of the membranes (manually wiping with a sponge) was performed at the time points indicated by the arrows in Fig. 1. The TMP in the Control-MBR (Fig. 1 (a)) rapidly increased. Although the physical cleaning substantially reduced the TMP, a rapid TMP increase was repeatedly observed in the Control-MBR. The rate of TMP increase obviously accelerated after 20 days of operation. As stated above, the concentrations of dissolved organic matter rapidly increased after 20 days of operation in the Control-MBR. It should be noted that the concentration of dissolved proteins doubled after 20 days of operation in the Control-MBR (Table 4). Dissolved polysaccharides and proteins in mixed liquor suspensions were assumed to be the major fractions of foulant in MBRs [28]. The increase in the concentrations of dissolved organic matter might have caused severe membrane fouling after 20 days of operation in the Control-MBR. The TMP in MBR-A (Fig. 1 (b)) further rapidly increased. To continue the operation, physical cleaning of the membrane needed to be frequently performed. Physical cleaning was very effective in the Control-MBR and MBR-A. This result means that the membrane fouling in these MBRs was mainly physically reversible fouling. In contrast, MBR-B and MBR-C could be operated for 30 days without any physical cleaning, and the TMP hardly increased during the operation (Fig. 1 (c), (d)). In MBR-C, an additional aerator was inserted to promote movement of the sponge carriers because mobility of the sponge carriers decreased during the operation. As a result, the aeration rate was slightly increased in the case of MBR-C. The net aeration rate in MBR-C was 17.3 L/min (2% increase). Membrane fouling could be effectively controlled by the granular or sponge carrier in MBR-B and MBR-C.

1 Fig. 1. Time course change of TMP in each MBR.

2

3 The fouled membranes were observed using SEM at the termination of the operation (Fig. 2).
 4 The membrane surface was fully covered by a cake structure in the Control-MBR and
 5 MBR-A (Fig. 2 (b), (c)). In contrast, the membrane surface was almost intact in MBR-B and
 6 MBR-C (Fig. 2 (d), (e)). Wang et al. [29] reported that a gel layer eventually developed even
 7 when routine aeration was carried out. In this study, however, the SEM observation clearly
 8 demonstrated that fluidizing carriers in the MBRs completely removed the cake/gel layer
 9 from the membrane surface.

10 Fig. 2. SEM images of the membrane surfaces at the termination of operation:

11 (a) virgin membrane, (b) Control-MBR, (c) MBR-A, (d) MBR-B and (e) MBR-C.

3.4 Filterability of the mixed liquor suspension in the four MBRs

Fig. 3 shows the filterability of mixed liquor suspensions in each MBR assessed by batch filtration experiments. The assessments were conducted at the termination of the 30-day operation. In MBR-B used with granular carriers, the filterability of the mixed liquor suspension was significantly low. In contrast, the filterability of the mixed liquor suspension in MBR-C used with sponge carriers was comparable to the Control-MBR. The concentrations of dissolved organic matter in the Control-MBR (after 20 days of operation) were higher than that in MBR-B; however, the filterability of the mixed liquor suspension in MBR-B was lower than that in the Control-MBR. Characteristics of SMP produced in MBR-B might have a property to cause severe deterioration of the filterability of mixed liquor suspension. The characteristics of the SMP produced in MBR-B might cause severe deterioration of the filterability of the mixed liquor suspension. Although conventional measurements of polysaccharides and proteins by colorimetric methods do not show differences in SMP characteristics, advanced analysis such as distribution of monosaccharides and amino acids may provide useful information [30,31]. Distribution of the monosaccharides in the SMP collected from each MBR is shown in Fig. 4. The compositions were fairly stable in each MBR and were different among the four MBRs. It should be noted that the percentage of fucose in MBR-B was higher than the other MBRs. Furthermore, a small percentage of galactose in the SMP collected from MBR-B was notable. This result indicated that the use of the granular carriers induced the different characteristics of SMP, which might have caused the deterioration of the filterability. The characteristics of the SMP produced in MBRs would change depending on the type of carriers, leading to different filterability of the mixed liquor suspensions. A further study to reveal the relationship between the filterability of the mixed liquor suspensions and the characteristics of the SMP is necessary. As stated before, the use of granular carriers in MBR-B could effectively mitigate the increase in TMP (Fig. 1(c)). The mechanical cleaning effect of the carriers can compensate for the deterioration of the filterability of the mixed liquor suspensions.

Fig. 3. Filterability of the mixed liquor suspensions in each MBR (n = 3).

Fig. 4. Distribution of the monosaccharides in the SMP from each MBR (n = 9).

^a n = 6; ^b n = 3.

3.5 Analysis of the fouled membrane

The filtration resistances of the fouled membrane analysed at the termination of the operation are shown in Fig. 5. Sodium hydroxide was used as the reagent for alkaline cleaning in this analysis. The fouled membranes after physical cleaning were soaked for 24 h at 30 °C in an alkaline solution (pH = 12). In Fig. 5, the degrees of physically irreversible fouling are shown by the difference between the filtration resistance of the virgin membrane and the filtration resistance of the fouled membrane after physical cleaning [32]. Irreversible fouling occurred in the four MBRs. It is shown that the development of irreversible fouling was significant in MBR-B. The authors have reported that the introduction of granular carriers into the MBR accelerated irreversible fouling [5]. The reason for this phenomenon was assumed to be that foulants easily accessed the membrane surface because of the absence of the cake/gel

layer, which eventually promoted irreversible fouling. Sponge carriers also prevented accumulation of the cake/gel layer on the membrane surface. Therefore, it is expected that foulants could easily access the membrane surface in MBR-C, leading to severe irreversible fouling. However, as shown in Fig. 5, the development of irreversible fouling in MBR-C was not significant. As described in section 3.2, the concentrations of the dissolved organic matter in MBR-C were lower than that in MBR-B, and foulants in MBR-C might be scarce. Although this could be one of the reasons for less irreversible fouling in MBR-C, it should be noted that the SMP concentration measured by conventional methods (e.g., TOC) does not correspond to the development of irreversible fouling in MBRs [33]. As shown in Fig. 4, the characteristics of the SMP produced in each MBR were different. SMP produced in MBR-C could have a low potential to cause irreversible fouling compared to that in MBR-B. In MBR-A, the membrane surface was covered by a cake/gel layer. However, the degree of irreversible fouling was significant and close to MBR-B. In addition, restoration of the permeability by alkaline cleaning was the poorest with the membrane collected from MBR-A. The foulants that caused irreversible fouling in MBR-A bound more tightly to the membrane than the foulants of the other MBRs. The characteristics of irreversible fouling were different in each MBR, which is probably explained by the differences in the SMP produced. The characteristics of the SMP produced in MBRs with different carriers should be investigated with advanced analyses [34,35], which may reveal the mechanisms behind why different carriers result in different degrees of irreversible fouling.

Fig. 5. The filtration resistances of the fouled membrane at the termination of operation.

4. Conclusions

The influence of different biofilm carriers on the operation and membrane fouling in MBRs was investigated in this study. Using rope or sponge carriers with MBR processes improved the removal efficiency of nitrogen. However, membrane fouling became very severe when rope carriers were used. Using granular or sponge carriers effectively mitigated the physically reversible fouling because of their mechanical cleaning effect on the membrane surface. On the other hand, the deterioration of the filterability of the mixed liquor suspensions was significant with the granular carriers, leading to the evolution of physically irreversible fouling. The characteristics of the SMP significantly varied depending on the types of carriers used, which could influence the filterability of mixed liquor suspensions and the development of irreversible membrane fouling in MBRs.

Acknowledgement

This work was financially supported by the Japan Society for the Promotion of Science (JSPS) Grant-in-Aid for JSPS Fellows (grant number: 26·1222) and Core Research of Evolutional Science & Technology (CREST) for “Innovative Technology and System for Sustainable Water Use” from the Japan Science and Technology Agency (JST).

References

- [1] L. Deng, W. Guo, H.H. Ngo, J. Zhang, S. Liang, S. Xia, Z. Zhang, J. Li, A comparison study on membrane fouling in a sponge-submerged membrane and a conventional membrane bioreactor, *Bioresour. Technol.* 165 (2014) 69–74.
- [2] B. Siembida, P. Cornel, S. Krause, B. Zimmermann, Effect of mechanical cleaning with granular material on the permeability of submerged membrane in the MBR process, *Water Res.* 44 (2010) 4037–4046.
- [3] X. Huang, C.-H. Wei, K.-C. Yu, Mechanism of membrane control by suspended carriers in a submerged membrane bioreactor, *J. Membr. Sci.* 309 (2008) 7–16.
- [4] L. Jin, S.L. Ong, H.Y. Ng, Fouling control mechanism by suspended biofilm carriers addition in submerged ceramic membrane bioreactors, *J. Membr. Sci.* 427 (2013) 250–258.

- [5] T. Kurita, K. Kimura, Y. Watanabe, The influence of granular materials on the operation and membrane fouling characteristics of submerged MBRs, *J. Membr. Sci.* 469 (2014) 292–299.
- [6] Q. Yang, J. Chen, F. Zhang, Membrane fouling control in a submerged membrane bioreactor with porous, flexible suspended carriers, *Desalination* 189 (2006) 292–302.
- [7] T. Kurita, K. Kimura, Y. Watanabe, Energy saving in the operation of submerged MBRs by the insertion of baffles and the introduction of granular materials, *Sep. Purif. Technol.* 141 (2015) 207–213.
- [8] S.J. Khan, S. Ilyas, S. Javid, C. Visvanathan, V. Jegatheesan, Performance of suspended and attached growth MBR systems in treating high strength synthetic wastewater, *Bioresour. Technol.* 102 (2011) 5331–5336.
- [9] Q. Liu, X.C. Wang, Y. Liu, H. Yuan, Y. Du, Performance of a hybrid membrane bioreactor in municipal wastewater treatment, *Desalination* 258 (2010) 143–147.
- [10] J. Lee, W.-Y. Ahn, C.-H. Lee, Comparison of the filtration characteristics between attached and suspended growth microorganisms in submerged membrane bioreactor, *Water Res.* 35 (2001) 2435–2445.
- [11] L. Rodríguez-Hernández, A.L. Esteban-García, I. Tejero, Comparison between a fixed bed hybrid membrane bioreactor and a conventional membrane bioreactor for municipal wastewater treatment: A pilot-scale study, *Bioresour. Technol.* 152 (2014) 212–219.
- [12] Y. Liu, Z. Liu, A. Zhang, Y. Chen, X. Wang, The role of EPS concentration on membrane fouling control: Comparison analysis of hybrid membrane bioreactor and conventional membrane bioreactor, *Desalination* 305 (2012) 38–43.
- [13] K. Sombatsompopa, C. Visvanathan, R. Ben Aim, Evaluation of biofouling phenomenon in suspended and attached growth membrane bioreactor systems, *Desalination* 201 (2006) 138–149.
- [14] S.J. Khan, Z.-U. Rehman, C. Visvanathan, V. Jegatheesan, Influence of biofilm carriers on membrane fouling propensity in moving biofilm membrane bioreactor, *Bioresour. Technol.* 113 (2012) 161–164.
- [15] B. Rafiei, F. Naeimpoor, T. Mohammadi, Bio-film and bio-entrapped hybrid membrane bioreactors in wastewater treatment: Comparison of membrane fouling and removal efficiency, *Desalination* 337 (2014) 16–22.
- [16] J. Hu, H. Ren, K. Xu, J. Geng, L. Ding, X. Yan, K. Li, Effect of carriers on sludge characteristics and mitigation of membrane fouling in attached-growth membrane bioreactor, *Bioresour. Technol.* 122 (2012) 35–41.

- [17] D.D. Trapani, G.D. Bella, G. Mannina, M. Torregrossa, G. Viviani, Comparison between moving bed-membrane bioreactor (MB-MBR) and membrane bioreactor (MBR) systems: Influence of wastewater salinity variation, *Bioresour. Technol.* 162 (2014) 60–69.
- [18] S. Yang, F. Yang, Z. Fu, R. Lei, Comparison between a moving bed membrane bioreactor and a conventional membrane bioreactor on membrane fouling, *Bioresour. Technol.* 100 (2009) 6655–6657.
- [19] Q.-Y. Yang, T. Yang, H.-J. Wang, K.-Q. Liu, Filtration characteristics of activated sludge in hybrid membrane bioreactor with porous suspended carriers (HMBR), *Desalination* 249 (2009) 507–514.
- [20] I. Ivanovic, T.O. Leiknes, The biofilm membrane (BF-MBR)—a review, *Desalin. Water Treat.* 37 (2012) 288–295.
- [21] OECD, OECD Guidelines for Testing Chemicals, Organization for Economic Cooperation and Development Paris, Guideline 302A & 303A, 1993.
- [22] M. Dubois, K.A. Gilles, J.K. Hamilton, P.A. Rebers, F. Smith, Colorimetric method for determination of sugars and related substances, *Anal. Chem.* 28 (1956) 350–356.
- [23] O.H. Lowry, N.J. Rosebrough, A.L. Farr, R.J. Randall, Protein measurement with the folin phenol reagent, *J. Biol. Chem.* 193 (1951) 265–275.
- [24] J.-P. Croué, M.F. Benedetti, D. Violleau, J.A. Leenheer, Characterization and copper binding of humic and nonhumic organic matter isolated from the South Platte River: evidence for the presence of nitrogenous binding site, *Environ. Sci. Technol.* 37 (2003) 328–336.
- [25] W. Yang, W. Syed, H. Zhou, Comparative study on membrane fouling between membrane-coupled moving bed biofilm reactor and conventional membrane bioreactor for municipal wastewater treatment, *Water Sci. Technol.* 69 (2014) 1021–1027.
- [26] S. Lyko, T. Wintgens, D. Al-Halbouni, S. Baumgarten, D. Tacke, K. Drensla, A. Janot, W. Dott, J. Pinnekamp, T. Melin, Long-term monitoring of a full-scale municipal membrane bioreactor – characterisation of foulants and operational performance, *J. Membr. Sci.* 317 (2008) 78–87.
- [27] D.J. Barker, D.C. Stuckey, A review of soluble microbial products (SMP) in wastewater treatment systems, *Water Res.* 33 (1999) 3063–3082.
- [28] A. Drews, Membrane fouling in membrane bioreactors – characterisation, contradictions, cause and cures, *J. Membr. Sci.* 363 (2010) 1–28.
- [29] Z. Wang, Z. Wu, X. Yin, L. Tian, Membrane fouling in a submerged membrane bioreactor (MBR) under sub-critical flux operation: Membrane foulant and gel layer characterization, *J. Membr. Sci.*, 325 (2008) 238–244.

- 1 [30] N. Yamato, K. Kimura, T. Miyoshi, Y. Watanabe, Difference in membrane fouling in
2 membrane bioreactors (MBRs) caused by membrane polymer materials, *J. Membr. Sci.* 280
3 (2006) 911–919.
- 4 [31] K. Kimura, T. Miyoshi, T. Naruse, N. Yamato, R. Ogyu, Y. Watanabe, The difference in
5 characteristics of foulants in submerged MBRs caused by the difference in the membrane flux,
6 *Desalination* 231 (2008) 268–275.
- 7 [32] K. Kimura, Y. Hane, Y. Watanabe, G. Amy, N. Ohkuma, Irreversible membrane fouling
8 during ultrafiltration of surface water, *Water Res.* 38 (2004) 3431–3441.
- 9 [33] K. Kimura, T. Naruse, Y. Watanabe, Changes in characteristics of soluble microbial
10 products in membrane bioreactors associated with different solid retention times: Relation to
11 membrane fouling, *Water Res.* 43 (2009) 1033–1039.
- 12 [34] K. Kimura, S.-I. Nishimura, R. Miyoshi, A. Hoque, T. Miyoshi, Y. Watanabe, Application
13 of glyco-blotting for identification of structures of polysaccharides causing membrane fouling
14 in a pilot-scale membrane bioreactor treating municipal wastewater, *Bioresour. Technol.* 179
15 (2015) 180–186.
- 16 [35] T. Miyoshi, T. Aizawa, K. Kimura, Y. Watanabe, Identification of proteins involved in
17 membrane fouling in membrane bioreactors (MBRs) treating municipal wastewater, *Int.*
18 *Biodeterior. Biodegrad.* 75 (2012) 15–22.