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New functional biocarriers for enhancing the performance of a hybrid moving bed biofilm reactor–membrane bioreactor system

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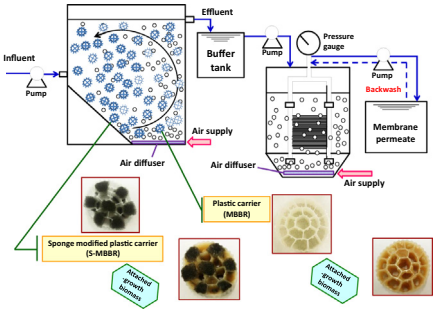
HIGHLIGHTS

- Plastic carriers were modified using sponge and then applied in MBBR.
- Sponge modified biocarriers could improve nutrient removal and effluent quality.
- The S-MBBR–MBR presented less membrane fouling and longer operative time.
- SMP in mixed liquor, R_C and R_P were reduced by the sponge modified biocarriers.
- Sponge modified biocarriers could enhance the treatability of the MBBR–MBR system.

GRAPHICAL ABSTRACT



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ABSTRACT

In this study, new sponge modified plastic carriers for moving bed biofilm reactor (MBBR) was developed. The performance and membrane fouling behavior of a hybrid MBBR–membrane bioreactor (MBBR–MBR) system were also evaluated. Comparing to the MBBR with plastic carriers (MBBR), the MBBR with sponge modified biocarriers (S-MBBR) showed better effluent quality and enhanced nutrient removal at HRTs of 12 h and 6 h. Regarding fouling issue of the hybrid systems, soluble microbial products (SMP) of the MBR unit greatly influenced membrane fouling. The sponge modified biocarriers could lower the levels of SMP in mixed liquor and extracellular polymeric substances in activated sludge, thereby mitigating cake layer and pore blocking resistances of the membrane. The reduced SMP and biopolymer clusters in membrane cake layer were also observed. The results demonstrated that the sponge modified biocarriers were capable of improving overall MBBR performance and substantially alleviated membrane fouling of the subsequent MBR unit.

1. Introduction

It has been widely accepted that biological nutrient removal (BNR) process is a preferable choice for simultaneous organic and nutrient removal during wastewater treatment. Currently, various BNR processes have been developed, including the five-stage

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Bardenpho process, the anoxic/oxic (A/O), the anaerobic/anoxic/oxic (A^2/O), the University of Cape Town (UCT) process, and attached biofilm reactors (Chen et al., 2011). Among them, the moving bed biofilm reactor (MBBR) is a cost-effective and efficient BNR technology, which can realize high-volume biofilm growth, high specific biomass activity, low headloss, no medium channelling and clogging, and inhibition of the excessive abrasive removal of slow growing microorganisms (Guo et al., 2010, 2012; Ødegaard, 1999).

During the operation process, characteristics of attached growth media play a key role in MBBR performance. In recent years, different kinds of media have been employed in MBBRs for wastewater treatment, including plastic media (e.g. suspended plastic biocarreriers, Kaldnes K1, K2, K3 and K5, Kaldnes biofilm Chip M, etc.), polyurethane foam, activated carbon (granular and powdered), natural occurring materials (e.g. sand, zeolite, diatomaceous earth, light expended clay aggregate, etc.), non-woven carriers, ceramic carriers, modified carriers (e.g. BIOCONS carrier, bioplastic-based moving bed biofilm carriers, polyvinyl alcohol-gel carrier, biodegradable polymer polycaprolactone carriers, etc.) and wood chips. The most popularly used carrier for MBBR is plastic media. A lab-scale MBBR containing 50% (filling ratio) of the Kaldnes biomedia K1 was operated by Aygun et al. (2008) for synthetic wastewater treatment. It was reported that the increase of the organic loading rate (6–96 g COD/m^2 d) caused the declined organic removal efficiency from 95.1% to 45.2%. Shore et al. (2012) used bench scale MBBRs with 50% fill of BioPortz™ media (high density polyethylene (HDPE)) to treat secondary treated effluent. They found that more than 90% of NH_4-N was eliminated from both synthetic and industrial wastewater at 35 and 40 °C by the MBBRs. Zhang et al. (2013) used a pilot-scale MBBR with suspended polyethylene (PE) bio-carriers having inclination angle of 60° (50% of working volume fraction) to treat the raw water polluted by NH_4-N at various temperatures (3.7–35.7 °C) and NH_4-N loadings (0.031–0.0473 g NH_4^+-N/m^2 d), achieving average removal of 71.4 ± 26.9%. However, the MBBR systems with plastic media generally do not present high T-N removal due to their limited denitrification capacity. Moreover, under aeration condition in MBBR, the strict anaerobic zone cannot be obtained for effective phosphorus release, which in turn decreases phosphorus removal efficiency (Zhuang et al., 2014). Other constrains include long start-up period required for biofilm growth on the plastic media and stabilizing system performance (Habouzit et al., 2014), as well as easy detachment of biofilm from the plastic media (Rafiei et al., 2014).

As a promising alternative media, sponge has attracted more and more interest, as it is low-cost material and can promote the rapid and stable attachment and growth of microorganisms on the carrier due to its high porosity (Ngo et al., 2008). Some recent studies have highlighted the effectiveness of sponge in MBBRs for organic and nutrient removal. The batch experiments conducted by Lim et al. (2011) showed that high concentrations of 8-mL polyurethane sponge cubes (2 × 2 × 2 cm, 40% (v/v)) induced good T-N removal of 84% in treating low COD/N ratio wastewater. It could enable high capacity of the moving bed sequencing batch reactors (MBSBRs) for nitrogen removal at low cost. Chu and Wang (2011) reported that the MBBR with sponge (20% filling ratio) at a hydraulic retention time (HRT) of 14 h showed high TOC and NH_4-N removal (90% and 65%, respectively). Feng et al. (2012) also pointed out that the aerobic MBBR with high polyurethane foam packing rate of 40% could remove average 80% of COD and 96.3% of NH_4-N for artificial sewage treatment at an HRT of 5 h.

This study focused on investigating new functional media (i.e. plastic carrier modified using sponge) developed at UTS for enhancing the treatment performance of MBBR system. Based on literature, it is the first development of this specific configuration of new biofilm carriers which have a combined structure of sponge and plastic carriers. This aims to improve the pollutant removal efficiency of MBBR while minimising membrane fouling of the subsequent membrane bioreactor (MBR) unit. The organic, nitrogen and phosphorus removals were elevated and compared between an MBBR with sponge modified plastic carriers (S-MBBR) and an MBBR with plastic carriers only. Both MBBRs were then coupled with membrane bioreactor (MBR) and the performance of two hybrid MBBR–MBR systems were also studied in terms of pollutant removal and membrane fouling.

2. Methods

2.1. Wastewater and media specifications

In this study, a synthetic wastewater with COD:N:P ratio of 100:5:1 was used to simulate primarily treated domestic wastewater, which was prepared with glucose, ammonium sulfate, potassium dihydrogen orthophosphate together with trace nutrients by dissolving in tap water. It gives dissolved organic carbon (DOC) of 100–130 mg/L, chemical oxygen demand (COD) of 330–360 mg/L, ammonium nitrogen (NH_4-N) of 12–15 mg/L, and orthophosphate of 3.3–3.5 mg/L. The pH was maintained at 7.0 by adding sodium carbonate or sulfuric acid on a daily basis.

The sponge modified plastic carrier was prepared by combining reticulated porous polyester-polyurethane sponge (Joyce Foam Products, Australia) with plastic carrier (namely Suspended Biological Filter, SBF® from Yixing City Yulong F.P. Co., Ltd., China). Each plastic carrier has the nominal diameter and length of 25 and 9 mm, respectively, with specific density of 950 kg/m^3, specific surface area of 500 m^2/m^3, and void ratio of 95%. The sponge (density of 28–30 kg/m^3, cell count of 90 cells/in (90 cells per 25 mm)) was cut into required size and fixed into alternate holes of the plastic carrier. The average weights of these two kinds of carriers were 1.20 ± 0.04 g per sponge modified plastic carrier and 1.08 ± 0.03 g per plastic carrier.

2.2. Experimental setup and operating conditions

Two batch-scale MBBR systems with effective working volume of 12 L were used and both MBBRs were filled with 20% of carriers (working volume fraction). The MBBR with fresh sponge modified plastic carriers (S-MBBR) and the MBBR with fresh plastic carriers (MBBR) were acclimatized for 15 days before operating in continuous mode at the flow rate of 16.7 mL/min, corresponding to a HRT of 12 h. The dissolved oxygen (DO) concentration was controlled in the range of 5.0–6.0 mg/L for both MBBRs. The low air flow rate could promote complete liquid–solid mixing, moderate media up/down motion, and limit the release of biomass from the media.

For the set-up of the hybrid systems, two 10-L submerged MBR units were employed to connect with the S-MBBR and the MBBR, hereafter referred to as S-MBBR–MBR and MBBR–MBR, respectively (Fig. 1). For the MBR unit, the membrane module used in this study was hollow fiber membrane which was made of polyethylene (PE) with hydrophilic coating having a surface area of 0.195 m^2 and a pore size of 0.1 μm. Infinite sludge retention time (SRT) was obtained without sludge waste. MBBR effluent was pumped into the MBR unit as the feed through a buffer tank. The membrane permeate was withdrawn from the membrane module by a suction pump at the filtration flux of 10.26 L/m^2 h to maintain the HRT at 5 h. There was a pressure gauge connected with membrane for measuring transmembrane pressure (TMP) value every day. Only two times/day backwash frequency with duration of 2 min/time was employed at a flow rate of 30.78 L/m^2 h. Chemical cleaning (1% hydrochloric acid, 2% citric acid, 0.4% sodium

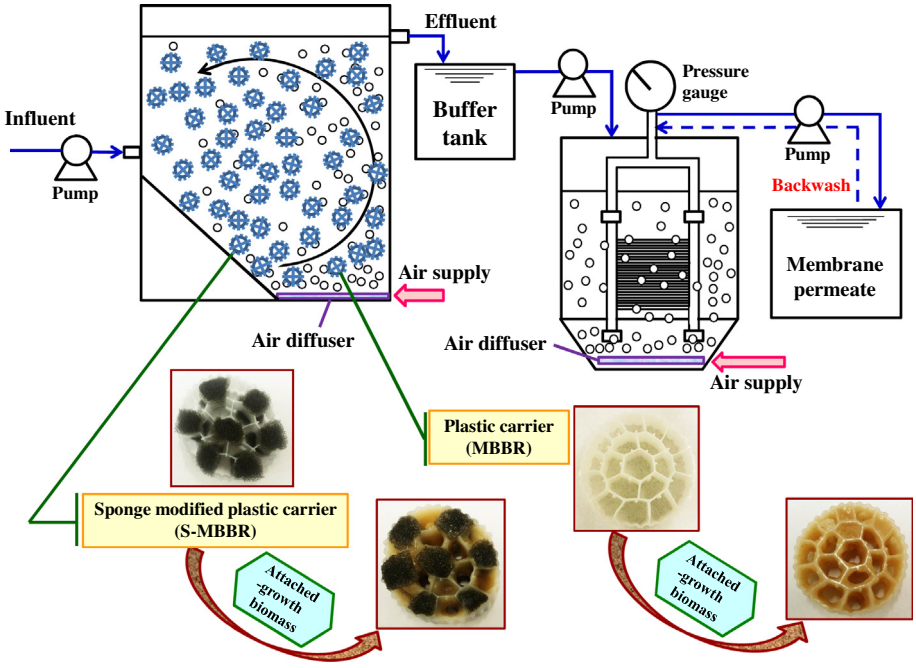

Fig. 1. Experimental set-up of the S-MBBR–MBR and the MBBR–MBR.

hypochlorite plus 4% sodium hydroxide for 6 h soaking, respectively) was conducted when terminating the experiments at TMP of 35.0 kPa.

The entire study period consisted of 5 phases according to different operating conditions as displayed in Table 1. Phase I (Day 0–15) is the acclimatization period for both MBBRs in batch mode until both systems reached relatively stable treatment performance. In Phase II (Day 16–30), both MBBRs were operated in continuous mode (flow rate of 16.7 mL/min). The stabilization of both MBBR systems was achieved within the first 30-day operation. During the experimental period, both MBBRs were operated at HRT of 12 h from Day 31 to 60 (Phase III). The HRT was then halved to 6 h from Day 61 to 90 (Phase IV) to match the flow rate requirement of the subsequent MBR unit (33.3 mL/min). Finally, the evaluation of two hybrid systems was conducted at Phase V at the HRTs of 6 h for the MBBR units and 5 h for the MBR units.

Table 1
Operating conditions at different phases over the entire experimental period.

Phase	Operational day	Systems	HRT (h)	Flow rate (mL/min)
I	0–15 (acclimatization period)	S-MBBR, MBBR	12	16.7
II	16–30 (stabilization period)	S-MBBR, MBBR	12	16.7
III	31–60	S-MBBR, MBBR	12	16.7
IV	61–90	S-MBBR, MBBR	6	33.3
V	91–175	S-MBBR–MBR	6 for MBBR unit, 5 for MBR unit	33.3
	91–122	MBBR–MBR		

S-MBBR: MBBR with sponge modified plastic carriers.
MBBR: MBBR with plastic carriers.
S-MBBR–MBR: Hybrid MBBR–MBR system with sponge modified plastic carriers.
MBBR–MBR: Hybrid MBBR–MBR system with plastic carriers.

2.3. Analysis methods

DOC analysis for samples was performed using the Analytikjena Multi N/C 2000. The Standard Methods were adopted for measurements of COD, attached-biomass and suspended sludge concentrations (including mixed liquor suspended solids (MLSS) and mixed liquor volatile suspended solids (MLVSS)) (APHA et al., 1998). The turbidity of the MBBR effluent was determined with 2100P Turbidimeter (HACH Company, USA). The photometric method called Spectroquant® Cell Test (NOVA 60, Merck) was used to quantify NH_4-N, NO_3-N, NO_2-N and PO_4-P. The extraction and analyses of extracellular polymeric substances (EPS) and soluble microbial products (SMP) in mixed liquor and cake layer in the MBR unit could refer to our previous study by Deng et al. (2014). Moreover, the extraction of biopolymer clusters (BPC) was performed based on the protocol of Sun et al. (2008). The extracted samples were analyzed for protein (EPS_P, SMP_P, BPC_P) and polysaccharide (EPS_C, SMP_C, BPC_C) concentrations, following the modified Lowry method (Sigma, Australia) and Anthrone-sulfuric acid method (Raunkjer et al., 1994), respectively.

According to the resistance-in-series model, membrane filtration characteristics were obtained using Eqs. (1) and (2) (Choo and Lee, 1996):

$$J = \Delta P/\mu R_T \quad (1)$$

$$R_T = R_M + R_C + R_P \quad (2)$$

where J is the permeate flux; ΔP is the TMP; μ is the viscosity of the permeate; R_T is the total resistance; R_M is the intrinsic membrane resistance; R_C is the cake resistance; and R_P is the pore blocking resistance.

3. Results and discussion

3.1. Treatment performance of the S-MBBR and the MBBR during start-up period

During the first 15-day operation (Phase I, start-up period), organic matter removal was obtained in both MBBRs with small

variations. The removal efficiency was 90.03 ± 3.68% for DOC and 89.57 ± 4.62% for COD in the S-MBBR. For the MBBR, DOC and COD removals were 88.90 ± 4.39% and 86.21 ± 5.03%, respectively. Nutrient removal of MBBRs exhibited significant changes with the elapsed time. In the S-MBBR, NH_4-N, T-N, and PO_4-P removals were 60.35 ± 10.21%, 60.32 ± 14.03%, and 63.92 ± 12.87%, respectively, while the MBBR presented less nutrient removals (54.15 ± 11.44%, 51.14 ± 13.46%, and 54.95 ± 13.42%, respectively). As biomass growth was initialized on the carriers during the acclimatization period, nutrient removals were low and unstable in both MBBRs.

From Day 16 to 30, both MBBRs approached steady state (Phase II). Better treatment performance was found in the S-MBBR (94.73 ± 3.85%, 93.26 ± 2.75%, 83.76 ± 4.06%, 75.26 ± 2.17% and 74.76 ± 3.93% for DOC, COD, NH_4-N, T-N and PO_4-P removals, respectively), compared to the MBBR (94.05 ± 4.76%, 92.03 ± 3.19%, 74.58 ± 5.19%, 59.90 ± 6.34% and 63.28 ± 6.28%, respectively). Additionally, the attached-biomass growth also reached steady state. The carriers in the S-MBBR contained more attached-growth biomass (0.1473 ± 0.0041 g MLSS/g and 0.1341 ± 0.0063 g MVLSS/g sponge modified plastic carrier) than those for the MBBR (0.0677 ± 0.0023 g MLSS/g and 0.0573 ± 0.0016 g MVLSS/g plastic carrier). For the plastic carrier, the biofilm was mainly developed on the outer surface of the carrier. As fresh sponge possesses large amount of pores, microorganisms can be entrapped into the pores and developed on both outer and inner surfaces of sponge (Guo et al., 2010). Hence, larger amount of biomass was attached onto the sponge modified plastic carrier as compared to that on the plastic carrier. For the suspended growth in the MBBRs, MLSS and MLVSS concentrations of mixed liquor in the S-MBBR remained at 0.251 ± 0.018 and 0.243 ± 0.016 g/L, respectively, which were similar to those in the MBBR (0.262 ± 0.031 and 0.250 ± 0.029 g/L, respectively).

3.2. Treatment performance of the S-MBBR and the MBBR during experimental period

After the steady state, the S-MBBR and the MBBR were operated at two HRTs of 12 h (Phase III) and 6 h (Phase IV) and the results are summarized in Table 2. At HRT of 12 h, stable DOC and COD removals of 95.63 ± 4.23% and 94.58 ± 5.06% were observed in the S-MBBR, respectively, which were higher than that for the MBBR (93.52 ± 3.25% for DOC removal and 91.27 ± 4.69% for COD removal). It suggested that the MBBR systems demonstrated good performance in organic matter removal. NH_4-N removal in the S-MBBR averaged at 83.46 ± 3.98%, which was approximately 10% higher than the MBBR. Nitrifying microorganisms (including ammonia oxidizing bacteria and nitrite oxidizing bacteria) could be kept by the biofilm on the media, thus giving high NH_4-N removal in the S-MBBR and the MBBR (Shore et al., 2012). Moreover, the results also showed that sponge modified plastic carriers could prevent more nitrifies being washed out with the effluent of the S-MBBR, leading to better NH_4-N removal. Nearly 14% higher T-N elimination achieved in the S-MBBR also implied that simultaneous nitrification and denitrification (SND) process took place, although DO in both MBBRs was maintained at relatively high levels of 5.0–6.0 mg/L in this study. This is due to that the oxic and the anoxic micro-zones could be formed at the outer layer and the inner layer of the biofilm, which was ascribed to DO concentration gradient within the biofilm of media owing to limited oxygen diffusion (Chu and Wang, 2011). Therefore, in the S-MBBR, as the declining DO levels along the inner depth of sponge also favoured the formation of the anoxic zone and permitted more effective denitrification process (Guo et al., 2008), the sponge modified biocarriers could enhance the SND process.

Table 2
Treatment performance of MBBRs at HRTs of 12 and 6 h during experimental period.

Removal efficiency (%)	Phase III		Phase IV	
	S-MBBR	MBBR	S-MBBR	MBBR
DOC	95.63 ± 4.23	93.52 ± 3.25	98.66 ± 1.10	95.89 ± 0.50
COD	94.58 ± 5.06	91.27 ± 4.69	97.52 ± 1.63	93.16 ± 1.45
NH_4-N	83.46 ± 3.98	72.75 ± 5.50	94.17 ± 1.62	81.30 ± 2.03
T-N	74.71 ± 2.06	60.15 ± 6.41	86.66 ± 1.15	71.80 ± 5.01
PO_4-P	70.63 ± 4.15	63.82 ± 6.01	84.52 ± 3.66	70.20 ± 1.89

During the operating period, phosphate can be taken up by phosphorus accumulating organisms in the oxic/anoxic zones (Monclús et al., 2010). As the SND process reduced NO_3-N content in both MBBRs (2.96 ± 0.68 and 3.82 ± 0.84 mg/L in the S-MBBR and the MBBR, respectively), the presence of less NO_3-N in the anoxic zones inside the biofilm resulted in effective PO_4-P release and thus promoted PO_4-P removal (Yuan et al., 2008; Yang et al., 2010). Furthermore, the sponge modified plastic carrier facilitated PO_4-P elimination by retaining more attached-growth biomass (Guo et al., 2008). Therefore, the S-MBBR obtained higher PO_4-P removal efficiency (70.63 ± 4.15%) than the MBBR (63.82 ± 6.01%).

At shortened HRT of 6 h, slightly higher DOC and COD removals in the S-MBBR (98.66 ± 1.10% and 97.52 ± 1.63%, respectively) and the MBBR (95.89 ± 0.50% and 93.16 ± 1.45%, respectively) were achieved. Both MBBRs also showed more desirable nutrient removal efficiencies. In addition, better effluent quality in terms of turbidity was observed at HRT of 6 h (17.14 ± 3.12 NTU for the S-MBBR and 56.35 ± 4.72 NTU for the MBBR), compared with higher effluent turbidity values obtained at HRT of 12 h (40.30 ± 3.67 NTU for the S-MBBR and 72.05 ± 4.82 NTU for the MBBR). At HRT of 12 h, the average food to microorganism (F/M) ratios were 0.07 kg BOD_5/kg MLVSS d for the S-MBBR and 0.17 kg BOD_5/kg MLVSS d for the MBBR. With decreased HRT of 6 h, F/M ratios increased up to 0.20 and 0.50 kg BOD_5/kg MLVSS d, respectively. At shorter HRT, F/M ratios in both MBBRs were within the normal range of the activated sludge processes (0.2–0.5 kg BOD_5/kg MLVSS d) (Javid et al., 2013). Thus, adequate substrate could be supplied for the microbial activities (including attached- and suspend-growth), leading to better treatment performance. On the other hand, at longer HRT, lower F/M ratio implied less substrate available for biomass in the reactors, which may cause the risk of sludge bulking and growing filamentous bacteria, thereby deteriorating effluent quality (Javid et al., 2013).

3.3. Performance of hybrid MBBR–MBR systems

3.3.1. Treatment performance

As shown in Fig. 2, the S-MBBR–MBR and the MBBR–MBR showed the excellent DOC removal (98.93 ± 0.89% and 96.64 ± 0.59%, respectively) and COD removal (98.27 ± 0.94% and 94.56 ± 1.06%, respectively). 96.06 ± 1.04% of NH_4-N, 85.60 ± 2.08% of T-N, and 84.08 ± 1.41% of PO_4-P were reduced by the S-MBBR–MBR, while the corresponding pollutant removals in the MBBR–MBR were found to be lower at 82.47 ± 1.88%, 69.59 ± 2.51%, and 68.83 ± 2.36% on average, respectively. It was clear that the MBBR unit could substantially eliminate pollutants in the hybrid systems.

3.3.2. Membrane fouling behavior

Fig. 3 shows the TMP variations for the MBBR–MBR and the S-MBBR–MBR with evolution of time. During the operation, TMP profile of the MBBR–MBR showed a rapid rise until TMP reached 35.0 kPa after 32-day operation, leading to a significantly higher fouling rate of 1.09 kPa/d. On the contrary, a gradual and progressive TMP increment was observed in the S-MBBR–MBR during first 78-day operation with initial TMP of 2.0 kPa and a sudden TMP jump from 20.0 to 35.0 kPa lasting for 7 days thereafter, resulting

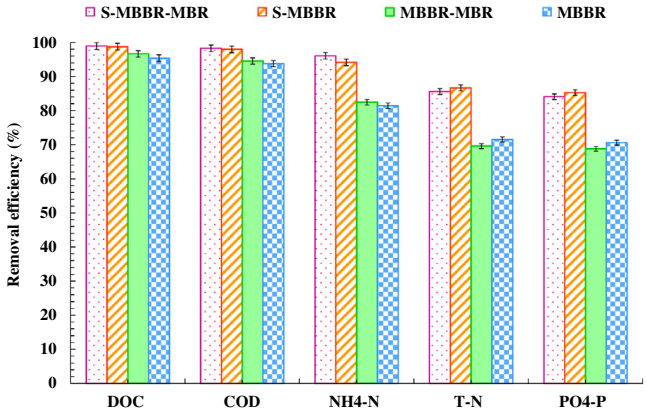

Fig. 2. DOC, COD, NH_4-N, T-N and PO_4-P removals in the S-MBBR–MBR, the S-MBBR, the MBBR–MBR, and the MBBR.

in a considerably lower fouling rate of 0.39 kPa/d. Thus, more effective membrane fouling mitigation for the S-MBBR–MBR was attributed to extended filtration duration and improved filterability. Moreover, fouling resistance for the fouled membrane in both hybrid systems was measured at the end of the experiment for further fouling analysis. As shown in Table 3, total fouling resistances (R_T) were 3.06×10^{12} and 1.42×10^{12} m^{-1} in the MBBR–MBR and the S-MBBR–MBR, respectively. Cake layer in the MBBR–MBR possessed a higher filtration resistance (R_C) of 1.29×10^{12} m^{-1}, while R_C for the S-MBBR–MBR was comparatively lower (0.47×10^{12} m^{-1}), which accounted for 42.16% and 33.10% of R_T, respectively. Pore blocking resistance (R_P) for the MBBR–MBR was higher than 3 times comparing with that for the S-MBBR–MBR, corresponding to 39.54% and 27.46% of R_T, respectively. The high importance of R_P on R_T in this study may be due to the fact that the MBR unit mainly contained solutes and colloids originating from MBBR effluent, giving rise to serious pore blocking (Defrance et al., 2000; Radjenović et al., 2008). Overall, the S-MBBR–MBR exhibited better membrane permeability by ameliorating pore blocking and cake layer formation.

It has been reported that EPS facilitated the formation of a cake layer and/or a highly hydrated gel layer containing microbial cells on membrane surface, which further prompted membrane pore blocking (Lin et al., 2014). In addition, SMP encouraged membrane pore blocking, and occupied the space among the particles of cake layer, resulting in a low porosity of cake layer (Domínguez et al., 2012). Figs. 4 and 5 display the levels of EPS_P and EPS_C of activated sludge, total SMP contents and SMP_P/SMP_C ratios in the

Table 3
Fouling resistance distribution in the MBBR–MBR and the S-MBBR–MBR.

Resistance distribution	MBBR–MBR		S-MBBR–MBR	
	m^{-1}	% of R_T	m^{-1}	% of R_T
Total	3.06×10^{12}		1.42×10^{12}	
Cake layer	1.29×10^{12}	42.16	0.47×10^{12}	33.10
Pore blocking	1.21×10^{12}	39.54	0.39×10^{12}	27.46
Clean membrane	0.56×10^{12}	18.30	0.56×10^{12}	39.44

R_T = total fouling resistance.

supernatant of mixed liquor in the MBR unit at different designated TMP values. Prior to a sudden TMP jump (20 kPa), EPS (EPS_P and EPS_C) of both MBBR–MBRs were at low values and presented slight difference. At TMP of 20 kPa, the notable differences of EPS_P and EPS_C levels between the MBBR–MBR (3.86 and 3.59 mg/L, respectively) and the S-MBBR–MBR (2.15 and 1.85 mg/L, respectively) were observed. When TMP reached the highest designated value of 35 kPa, EPS_P and EPS_C contents in the MBBR–MBR reached the highest values of 7.56 and 7.62 mg/L, respectively, which were almost 3 times of the corresponding values for the S-MBBR–MBR (2.31 and 2.85 mg/L, respectively). When TMPs were below 20 kPa, SMP gradually increased from 5.29 to 13.05 mg/L in the MBBR–MBR, while those values maintained at a lower range of 2.99–7.06 mg/L in the S-MBBR–MBR. During the severe membrane fouling period (TMP from 20 to 35 kPa), SMP levels in the MBBR–MBR rose dramatically from 17.58 to 25.86 mg/L. In contrast, the S-MBBR–MBR possessed considerably less SMP and exhibited more stable SMP levels between 7.52 and 9.93 mg/L. The results indicated that total concentrations of SMP were substantially higher than those of EPS in both hybrid systems. Additionally, higher EPS and SMP levels in the MBR unit of the MBBR–MBR were ascribed to higher biomass growth rate in the MBR unit (0.029 g MLSS/L d) as compared to that (0.010 g MLSS/L d) of the S-MBBR–MBR. Hence, SMP made a greater contribution to membrane fouling development in the MBBR–MBR. Besides, SMP_P/SMP_C ratios in the MBBR–MBR (0.66 ± 0.15) were always lower than those in the S-MBBR–MBR (1.00 ± 0.24) at all the designated TMPs. Furthermore, SMP_C could exacerbate irreversible fouling and induce severe pore blocking and gel layer formation (Jermann et al., 2007). Thus, considerably larger amounts of SMP with lower SMP_P/SMP_C ratio of mixed liquor and higher concentrations of EPS of activate sludge were responsible for the elevated R_C and R_P of the MBBR–MBR.

The extracted EPS, SMP and BPC from the cake layer were also investigated and characterized by their compositions (including polysaccharides and proteins) (Table 4). Both hybrid MBBR–MBR

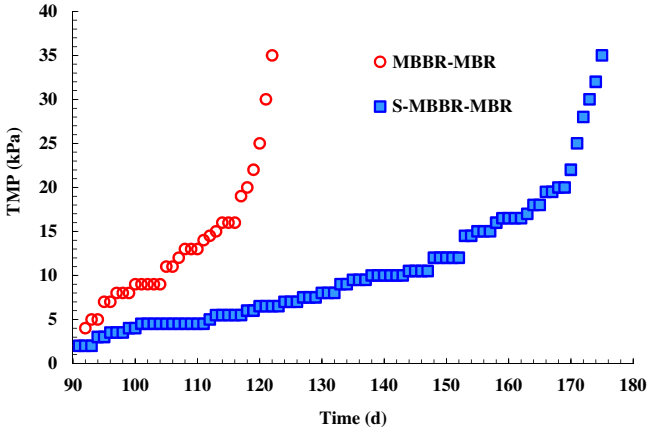

Fig. 3. TMP development profile for the MBBR–MBR and the S-MBBR–MBR.

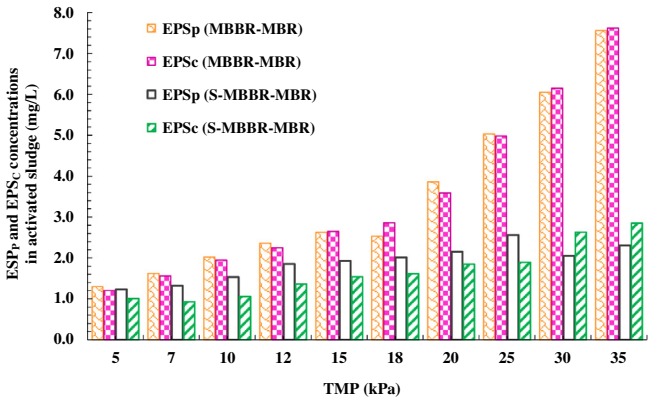

Fig. 4. Variations of EPS_P and EPS_C concentrations of activated sludge in the MBR unit at different TMPs.

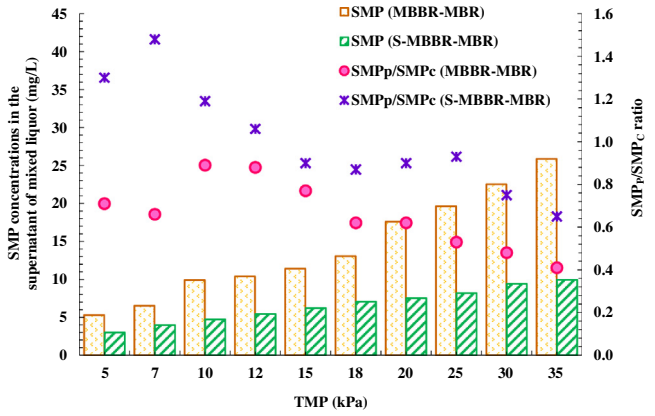

Fig. 5. Variations of SMP concentrations and SMP_P/SMP_C ratios of mixed liquor in the MBR unit at different TMPs.

Table 4
The compositions of bound EPS, SMP and BPC in membrane cake layer.

Reactors	EPS (mg/g cake layer)		SMP (mg/g cake layer)		BPC (mg/g cake layer)	
	EPS_P	EPS_C	SMP_P	SMP_C	BPC_P	BPC_C
S-MBBR–MBR	2.69	1.25	4.13	2.62	8.25	5.73
MBBR–MBR	3.02	1.66	8.63	5.62	15.27	12.16

systems had similar EPS levels (including EPS_P and EPS_C) at 4.68 and 3.94 mg/g cake layer for the MBBR–MBR and the S-MBBR–MBR, respectively. Cake layer for the MBBR–MBR was characterized by higher SMP_P and SMP_C levels than those for the S-MBBR–MBR. BPC_P and BPC_C contents for the MBBR–MBR were 15.27 and 12.16 mg/g cake layer, respectively, whereas those values remarkably decreased for the S-MBBR–MBR, obtaining 8.25 and 5.73 mg/g cake layer, respectively. Hence, cake layer formation for the MBBR–MBR was mainly caused by the accelerated growth of SMP (SMP_P and SMP_C) and BPC (BPC_P and BPC_C) within sludge cake, leading to higher R_C. Moreover, as sponge on the carriers could positively modify the characteristics of suspended biomass through adsorption and biodegradation of attached-biomass of sponge (Deng et al., 2014), it also contributed to the lower SMP and BPC values in the S-MBBR–MBR. In addition, higher drag force due to faster TMP increment in the MBBR–MBR might also enhance the growth of SMP and BPC on membrane surface, which encouraged the development of cake layer, further causing SMP generation by cell lysis and endogenous decay inside the bio-cake layer (Drews, 2010). These results again highlighted the significance of SMP on membrane fouling in the MBBR–MBR.

4. Conclusions

This study evaluated the feasibility and performance of sponge modified plastic carriers in both MBBR and MBBR–MBR systems. Compared to MBBR using plastic carriers, sponge modified biocarriers could not only enhance overall organic and nutrient removal efficiencies, but also prolong the operative time of the hybrid MBBR–MBR system due to efficient fouling reduction. The MBBR–MBR with sponge modified biocarriers exhibited lower SMP levels in mixed liquor with higher SMP_P/SMP_C ratio, as well as less pore blocking and cake layer resistances. Therefore, the sponge modified biocarriers could be a promising solution to improve the treatability of the MBBR–MBR system.

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