



## A comparison study on membrane fouling in a sponge-submerged membrane bioreactor and a conventional membrane bioreactor



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### HIGHLIGHTS

- Less SMP and bound EPS in activated sludge in the SSMBR induced lower  $R_c$  and  $R_p$ .
- Lower biomass growth and sludge viscosity contributed to lower  $R_c$  in the SSMBR.
- Larger sludge flocs, higher zeta potential and RH led to lower  $R_T$  in the SSMBR.
- Sponge could prevent pore blocking and cake layer formation.
- Sponge addition could reduce  $SMP_c$  and  $EPS_c$  through adsorption and biodegradation.

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### ABSTRACT

This study compared membrane fouling in a sponge-submerged membrane bioreactor (SSMBR) and a conventional membrane bioreactor (CMBR) based on sludge properties when treating synthetic domestic wastewater. In the CMBR, soluble microbial products (SMP) in activated sludge were a major contributor for initial membrane fouling and presented higher concentration in membrane cake layer. Afterwards, membrane fouling was mainly governed by bound extracellular polymeric substances (EPS) in activated sludge, containing lower proteins but significantly higher polysaccharides. Sponge addition could prevent cake formation on membrane surface and pore blocking inside membrane, thereby alleviating membrane fouling. The SSMBR exhibited not only less growth of the biomass and filamentous bacteria, but also lower cake layer and pore blocking resistance due to lower bound EPS concentrations in activated sludge. Less membrane fouling in SSMBR were also attributed to larger particle size, higher zeta potential and relative hydrophobicity of sludge flocs.

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### 1. Introduction

In the past decades, membrane bioreactor (MBR) has emerged as a considerably alternative to the conventional activated sludge treatment system for water reclamation and reuse. This technology has some superior merits, such as high effluent quality, small footprint, complete liquid–solid separation, high biomass content, absolute control of sludge retention time (SRT) and hydraulic retention time (HRT), and low sludge production (Guo et al., 2009). However, membrane fouling, especially biofouling, is the most obstacle in wide application of the MBR technology.

Generally, biofouling is referred to as undesirable accumulation of microorganisms at a phase transition interface, which may occur by deposition, growth and metabolism of bacteria cells or flocs on the membranes (Guo et al., 2012). As one of the most serious operational problems in membrane applications, biofouling causes severe flux decline, reduces membrane efficiency, increases membrane replacement and operational and maintenance costs.

Various strategies have been employed to reduce membrane fouling in the MBRs. Ngo and Guo (2009) found that an aerated submerged MBR (SMBR) system with addition of a very low-dose green bioflocculant (GBF) could achieve near zero membrane fouling after 70 days of operation as well as less backwash frequency. A chemical cleaning-in-place (CIP) was investigated by Wei et al. (2011) in a long-term operation of pilot-scale submerged MBR

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for municipal wastewater treatment. They reported that the chemical CIP, in both transmembrane pressure (TMP) controlling mode and time controlling mode, effectively removed the fouling in terms of membrane pore blockage and gel layer caused by colloids and soluble organic substances. Wu and He (2012) suggested that the low irreversible fouling was found in the cyclic aeration mode, which could be ascribed to the floc destruction and re-flocculation processes. During the short high aeration period, the preservation of the strong strength bonds within activated sludge flocs caused less release of soluble and colloidal material in the supernatant. The weak strength bonds damaged in the high aeration period could be recovered in the re-flocculation process in the low aeration period.

In addition, using biomass carriers (e.g. plastic media, powdered activated carbon (PAC), sponge) in MBR is an effective and promising method to control membrane fouling. Jin et al. (2013) suggested that biomass flocs were less easily broken up with addition of relatively light and large-sized suspended carriers (AnoxKaldnes, K1 carriers) in ceramic SMBR. Moreover, both extracellular polymer substances (EPS) and soluble microbial products (SMP) were lower in the SMBR with carriers than those in the SMBR without carriers. Ng et al. (2013) indicated that higher concentration of fresh PAC in the SMBR could provide better simultaneous adsorption, decomposition, and biodegradation effects for the reduction of fouling components in the supernatant of the mixed liquor such as EPS, fine colloids and planktonic cells. As an idea attached growth media, sponge has also exhibited excellent performance during biological treatment due to its advantages of high internal porosity and specific surface area, high stability to hydrolyses, light weight and low cost (Ngo et al., 2006). When employing in MBRs, it can act as a mobile carrier for active biomass, reduce cake layer formation on the membrane surface and retain microorganisms by incorporating both their attached growth and suspended growth (Ngo et al., 2008). Guo et al. (2008) investigated the effects of sponge addition on sustainable flux and membrane fouling. They found that compared to SMBR alone, the suspended sponge cubes in the sponge-submerged membrane bioreactor (SSMBR) with sponge volume fraction of 10% could significantly reduce the membrane fouling as well as improve sustainable flux by 2 times. Nguyen et al. (2012) also confirmed that SSMBR had lower TMP development than that of conventional SMBR during primary effluent treatment. Meanwhile, SSMBR could maintain good microbial activity and constant sludge volume index value.

Overall, previous studies have highlighted the advantages of sponge addition in MBRs for improving treatment performance as well as membrane fouling reduction in terms of sustainable flux or permeate flux. However, the effects of sponge on sludge characteristics and membrane fouling have yet to be investigated in MBR systems. Therefore, a comparison study was conducted to evaluate the performance of a SSMBR and a conventional MBR (CMBR) based on sludge characteristics, such as zeta potential, apparent viscosity, relative hydrophobicity (RH), EPS and SMP. The cake layer formation on membrane surface was also analysed.

## 2. Methods

### 2.1. Wastewater

The experiments were conducted using a synthetic wastewater to avoid any fluctuation in the feed concentration and provide a continuous source of biodegradable organic pollutants such as glucose, ammonium sulphate and potassium dihydrogen orthophosphate. It was used to simulate domestic wastewater just after primary treatment. The synthetic wastewater has dissolved organic carbon (DOC) of 100–130 mg/L, chemical oxygen demand

(COD) of 330–360 mg/L, ammonium nitrogen (NH<sub>4</sub>-N) of 12–15 mg/L and orthophosphate (PO<sub>4</sub>-P) of 3.3–3.5 mg/L. NaHCO<sub>3</sub> or H<sub>2</sub>SO<sub>4</sub> was used to adjust pH to 7.

### 2.2. Experimental setup and operating conditions

A SSMBR and a CMBR with the same effective working volume were operated in parallel to compare the performance and membrane fouling behaviour. For each MBR, a polyvinylidene fluoride (PVDF) hollow fiber membrane module with a pore size of 0.2 μm and surface area of 0.1 m<sup>2</sup> was used. Both MBRs were filled with sludge from a local Wastewater Treatment Plant and acclimatised to synthetic wastewater. They were started with identical seeding activated sludge with similar initial sludge concentration (7.03 g/L for SSMBR, 6.98 g/L for CMBR). No sludge was withdrawn from both MBRs. The reticulated porous polyester-polyurethane sponge (PUS) was used in SSMBR system. The PUS has density of 28–45 kg/m<sup>3</sup> and cell count of 45 cells/in (45 cells per 25.4 mm). The dimensions of the sponge cubes are 10 mm, 10 mm, and 10 mm in length, width and thickness, respectively. The sponge volume fraction was 10% in the SSMBR in this study, which was determined according to previous study of Guo et al. (2008). Before running the experiments, the sponge cubes were acclimatised to synthetic wastewater for 25 days. Synthetic wastewater was pumped into the reactor using a feeding pump to control the feed rate while the effluent flow rate was controlled by a suction pump. A pressure gauge was used to measure the TMP and a soaker hose air diffuser was used to maintain air flow rate at 9 L/min. The filtration flux of both MBRs was kept constant at 10 L/m<sup>2</sup> h by adopting a suction cycle of 59-min on and 1-min off (relaxation). For chemical cleaning of the membrane, the membrane was soaked in chemical solutions using the three following steps: 6 h in 0.5% citric acid, 6 h in 0.4% sodium hydroxide, 6 h in 0.8% sodium hypochlorite.

### 2.3. Analysis methods

DOC of the influent and effluent was measured using the Analytikjena Multi N/C 2000. The analysis of COD was according to Standard Methods (APHA, AWWA, WEF, 1998). NH<sub>4</sub>-N and PO<sub>4</sub>-P were measured by photometric method called Spectroquant<sup>®</sup> Cell Test (NOVA 60, Merck).

Fouling resistance was measured through various fluxes with distilled water at the end of the experiment. The resistance-in-series model was applied to evaluate membrane filtration characteristics by using Darcy's law. The model was expressed as follows (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;  $\Delta P$  is the TMP;  $\mu$  is the viscosity of the permeate;  $R_T$  is total resistance;  $R_M$  is the intrinsic membrane resistance;  $R_C$  is the cake resistance; and  $R_P$  is the pore blocking resistance.

At the end of the experiment, the membrane was taken out from the bioreactor. Cake layer on membrane surface was collected and then dissolved in 30 mL of distilled water. The extraction procedures and analysis methods of EPS and SMP of cake layer were in the same manner as described below. The EPS extraction protocol was modified from Frølund et al. (1996). 30 mL of mixed liquor were taken from the MBRs and then centrifuged at 3000 rpm for 30 min. After that, the supernatant was centrifuged at 3000 rpm for 30 min and filtered through 0.45 μm Phenex-NY (Nylon) syringe filter to obtain SMP. The pellets remaining in the centrifuge tube were suspended in phosphorus buffer solution up to 30 mL,

and then mixed with cation exchange resin for 2 h at 900 rpm. Extracted EPS were harvested by filtering the resin and liquid mixture through 1.2  $\mu\text{m}$  Phenex-GF (Glass fiber) syringe filter. In this study, the extracted samples were analysed for proteins (EPS<sub>P</sub> and SMP<sub>P</sub>) and polysaccharides (EPS<sub>C</sub> and SMP<sub>C</sub>) concentrations using modified Lowry method (Sigma, Australia) and Anthrone-sulphuric acid method, respectively.

The apparent viscosity and the zeta potential of mixed liquor were measured by Brookfield Viscometer M/OO-151-E0808 and Zetasizer Nano ZS (Malvern Instruments, UK), respectively. The relative hydrophobicity (RH) is the tendency of adherence of sludge flocs to hydrocarbon (*n*-hexane in this study) and was measured following the method by Ji et al. (2010). The equation RH (%) =  $(1 - \text{MLSS}_e/\text{MLSS}_i) \times 100\%$  was used to calculate RH, where MLSS<sub>e</sub> is the MLSS concentration in the aqueous phase after emulsification and MLSS<sub>i</sub> is the initial MLSS concentration of the sample. The difference between MLSS<sub>i</sub> and MLSS<sub>e</sub> is hydrocarbon phase and the concentration of sludge flocs adhering to *n*-hexane, indicating the hydrophobicity of sludge flocs. The images of sludge particles obtained by the Olympus System Microscope Model BX41 (Olympus, Japan) were acquired as jpg. format. Thereafter, the images were analysed with Image-Pro Plus software to obtain particle size distribution of sludge flocs.

### 3. Results and discussion

#### 3.1. The performance of SSMBR and CMBR

Table 1 summarises the removal efficiencies of DOC, COD, PO<sub>4</sub>-P, NH<sub>4</sub>-N and total nitrogen (TN) in SSMBR and CMBR during the operation period. As shown in Table 1, more than 90% of organic removal was obtained in both SSMBR and CMBR. SSMBR showed higher performance for removing NH<sub>4</sub>-N (>70%) and PO<sub>4</sub>-P (>60%), while around 60% of NH<sub>4</sub>-N and 30% of PO<sub>4</sub>-P were removed in the CMBR. Higher NH<sub>4</sub>-N removal in the SSMBR could be attributed to the enhanced population of ammonium oxidation bacteria on the acclimatised sponge during acclimatisation period (Nguyen et al., 2012). As sponge could provide the anoxic condition around the surface of the sponge and the anaerobic condition inside the sponge, the SSMBR achieved a higher removal efficiency of PO<sub>4</sub>-P (Guo et al., 2008).

Fig. 1 depicts the time course of TMP increase in both SSMBR and CMBR. Both MBRs demonstrated significant difference in TMP profiles. TMP in the SSMBR was maintained at 2.0 kPa up to 90 days. In the CMBR, TMP gradually increased from 5.0 kPa to 7.0 kPa until day 6, followed by a rapid TMP rise. After 35 days, the TMP reached 31.0 kPa, suggesting chemical cleaning should be conducted for the membrane. These results indicated that sponge addition could significantly mitigate membrane fouling, which is further discussed in details in Section 3.5.

#### 3.2. Mixed liquor suspended solids (MLSS) concentration and apparent viscosity

During the experimental period, sludge concentration kept increasing in both MBRs due to no sludge withdrawal. MLSS concentrations were  $11.50 \pm 4.52$  g/L and  $9.41 \pm 2.38$  g/L in the CMBR and SSMBR after 35 and 90 days of operation, respectively.

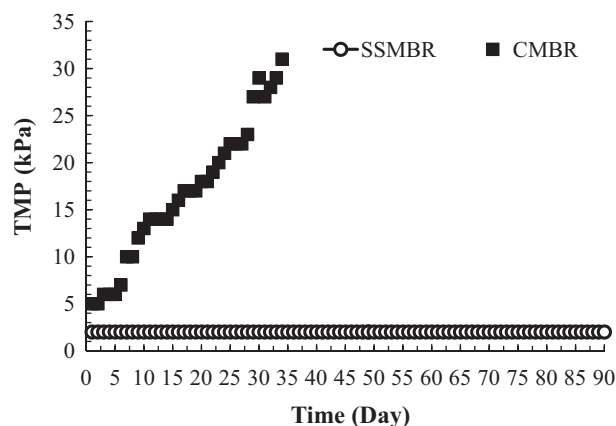


Fig. 1. TMP profile for SSMBR and CMBR.

The lower MLSS concentration in the SSMBR might be attributed to the fact that sponge addition could balance the microorganism growth in suspended activated sludge as well as on and inside the porous sponge cubes (Ngo et al., 2006). It was found that there is an exponential relationship between MLSS concentration and sludge viscosity (Reid et al., 2008). In this study, sludge viscosity was higher ( $3.30 \pm 0.50$  mPa s) in the CMBR than that ( $2.60 \pm 0.40$  mPa s) in the SSMBR, demonstrating that higher sludge viscosity was attributed to higher MLSS concentration. In addition, it has been reported that the sludge flocs with excess filamentous bacteria showed high viscosity due to presence of high EPS concentration (Meng et al., 2006a). Overgrowth of filamentous bacteria was found in the CMBR on day 14, whereas there were less filamentous bacteria in the SSMBR until 83 days, which revealed that higher sludge viscosity in the CMBR was also due to abundance of filamentous bacteria. Similar observations were also recorded by Meng et al. (2007) who suggested that sludge viscosity was influenced by MLSS concentration, EPS and filamentous bacteria.

#### 3.3. Zeta potential, relative hydrophobicity (RH) and particle size distribution

It has been demonstrated that the flocculation ability of sludge flocs is affected by their hydrophobicity and surface charge, which positively influences the hydrophobic interaction and electrostatic repulsion, respectively (Liao et al., 2001; Mikkelsen and Keiding, 2002). In this study, activated sludge in the SSMBR had higher zeta potential ( $-6.85 \pm 3.65$  mV) and higher RH ( $81.00 \pm 7.80\%$ ) than those in the CMBR (zeta potential of  $-10.50 \pm 4.50$  mV, RH of  $63.13 \pm 13.60\%$ ). The results indicated that there might be a positive relationship between surface charge (zeta potential) and hydrophobicity of activated sludge. Additionally, Meng et al. (2006a) reported that excess filamentous bacteria could prevent the agglomeration of floc particles by producing a bridge lattice due to the generation of abundant filaments from the flocs into the bulk solution. Results of particle size distribution in this study showed that larger sludge flocs (20–50  $\mu\text{m}$ ) were found in the SSMBR than those in the CMBR (10–40  $\mu\text{m}$ ). This suggested that

Table 1

Removal efficiencies of DOC, COD, PO<sub>4</sub>-P, NH<sub>4</sub>-N and TN in SSMBR and CMBR during the operation period.

Reactors	DOC (%)	COD (%)	PO <sub>4</sub> -P (%)	NH <sub>4</sub> -N (%)	TN (%)
SSMBR	94.74 ± 5.49	93.53 ± 4.46	63.57 ± 5.32	74.35 ± 3.22	53.28 ± 2.16
CMBR	94.17 ± 7.32	91.95 ± 6.53	27.22 ± 6.18	58.14 ± 6.13	37.20 ± 4.58

activated sludge had better flocculation ability in the SSMBR, which might be due to higher RH and zeta potential of sludge flocs as well as the presence of less filamentous bacteria.

### 3.4. Bound EPS and SMP in activated sludge

Normally, polysaccharides and proteins are considered as the major fractions of EPS and SMP that contribute to fouling (Guo et al., 2012). Tables 2 and 3 exhibit compositions of mixed liquor's SMP and bound EPS in the SSMBR and CMBR. The CMBR demonstrated higher SMP concentrations (around 2–3 times) within 7-day run. The protein concentrations (SMP<sub>p</sub>) were similar for both MBRs, while significantly higher polysaccharide concentrations (SMP<sub>c</sub>) were observed in the CMBR, suggesting higher fouling propensity of the CMBR. Although activated sludge of both MBRs had similar bound EPS concentrations, slightly higher protein concentrations (EPS<sub>p</sub>) but significantly lower polysaccharide concentrations (EPS<sub>c</sub>) were obtained in the CMBR. After 7 days of operation, the SMP concentrations (including SMP<sub>p</sub> and SMP<sub>c</sub>) of both MBRs presented minor difference. On the other hand, bound EPS concentrations (12.3–24.6 mg/L) in the CMBR were higher than those in the SSMBR (12.2–17.3 mg/L), with lower protein concentrations (EPS<sub>p</sub>) but significantly higher polysaccharide concentrations (EPS<sub>c</sub>). In this study, increase of sludge concentration under infinite SRT condition induced the decrease in food to microorganism (F/M) ratio (0.1–0.2 d<sup>-1</sup>). As a consequence, both MBRs were fed with limited available substrate, which could cause more cell lysis and cell hydrolysis, thereby releasing EPS and SMP in activated sludge (Yigit et al., 2008). Moreover, the excess growth of filamentous bacteria could produce more SMP, resulting in severe fouling (Pan et al., 2010). Therefore, the CMBR exhibited more serious fouling compared with the SSMBR. In the SSMBR, it was obvious that sponge addition could reduce SMP<sub>c</sub> during the first 7-day run and EPS<sub>c</sub> afterwards by the means of adsorption onto sponge as well as biodegradation by attached biomass of the sponge.

It has been reported that large quantity of EPS in activated sludge increased floc strength by polymer entanglement, thereby

increasing the extent of sludge flocs agglomeration (Mikkelsen and Keiding, 2002). However, in this study, lower EPS concentration but larger particles were observed in the SSMBR, pointing out that the flocculation ability of sludge flocs may not only depend on EPS concentration. Lee et al. (2003) found that the ratio of proteins to polysaccharides (PN/PS ratio) in EPS was important in controlling the hydrophobicity and surface charge of sludge flocs. Table 3 shows that a significantly higher PN/PS ratio in bound EPS was found in the SSMBR after 7 days operation. Higher RH of activated sludge in the SSMBR proved that higher EPS<sub>p</sub> concentration increased the hydrophobicity of sludge flocs by providing amino acids with more hydrophobic side groups, while lower EPS<sub>c</sub> concentration contributed to less hydrophilic nature of sludge. Moreover, the amino groups in EPS<sub>p</sub> containing positive charges neutralized some of negatively charged activated sludge, thereby inducing higher zeta potential of sludge flocs in the SSMBR (Lee et al., 2003; Liao et al., 2001). Thus, PN/PS ratio in bound EPS could positively influence hydrophobicity and zeta potential of activated sludge, thereby having an impact on the agglomeration ability of the flocs.

### 3.5. Membrane fouling behaviour

Results of fouling resistance showed that the CMBR had a higher total resistance ( $R_T$ ) ( $5.47 \times 10^{12} \text{ m}^{-1}$ ) than that of the SSMBR ( $2.56 \times 10^{12} \text{ m}^{-1}$ ). The clean membrane resistance ( $R_M$ ) were the same ( $1.71 \times 10^{12} \text{ m}^{-1}$ ) for both MBRs. Higher cake layer resistance ( $R_C$ ) was found for the CMBR than that for the SSMBR, corresponding to  $3.04 \times 10^{12} \text{ m}^{-1}$  and  $0.85 \times 10^{12} \text{ m}^{-1}$ , respectively. Moreover, pore blocking resistance ( $R_p$ ) for the CMBR was notably higher.  $R_p$  of the CMBR accounted for about 20% of  $R_T$ , whereas there was no  $R_p$  in the SSMBR. These results suggested that cake layer formation was one of the main factors contributing to membrane fouling. Furthermore, sponge could alleviate membrane fouling not only by preventing pore blocking but also by reducing cake layer formation. Some researchers (Jamal Khan et al., 2012; Yang et al., 2006) have reported the similar findings that  $R_C$  was major fraction of  $R_T$  and sponge addition could reduce  $R_C$ .

As discussed in Section 3.2, activated sludge in both MBRs possessed different properties, which were correlated with membrane fouling potential as well as fouling resistance. Higher MLSS concentration could lead to formation of a sticky cake layer on membrane surface due to higher sludge viscosity (Itonaga et al., 2004). Additionally, the sludge flocs with abundance of filamentous bacteria would more easily deposit on membrane surface due to its high viscosity, causing the formation of a non-porous cake layer (Meng et al., 2006a). Therefore, it could be noted that higher MLSS concentration and overgrowth of filamentous bacteria contributed to formation of sticky and non-porous cake layer, giving rise to higher  $R_C$  in the CMBR. Being the major fraction of the total fouling resistance, the cake layer was analysed with respect to EPS and SMP (including polysaccharides and proteins). Fig. 2 shows the compositions of EPS and SMP in the cake layer on membrane surface for both SSMBR and CMBR. Bound EPS concentrations were similar for the SSMBR (15.0 mg/(L g cake layer)) and the CMBR (13.9 mg/(L g cake layer)). However, higher concentrations of SMP<sub>c</sub> and SMP<sub>p</sub> (14.4 and 15.5 mg/(L g cake layer), respectively) were obtained for the CMBR, while SMP<sub>c</sub> and SMP<sub>p</sub> of the cake layer were comparatively lower for the SSMBR (9.8 and 7.1 mg/(L g cake layer), respectively). These results elucidated that higher  $R_C$  in the CMBR was mainly caused by SMP (including SMP<sub>c</sub> and SMP<sub>p</sub>) on membrane surface. At high TMP, more SMP<sub>c</sub> and SMP<sub>p</sub> could be adsorbed and/or attached onto membrane surface due to the high drag force provided by permeate pump. On contrary, sponge addition effectively reduced SMP<sub>c</sub> and SMP<sub>p</sub> in cake layer on membrane surface. Apart from adsorption of SMP<sub>c</sub>

**Table 2**

SMP compositions and total SMP concentrations of mixed liquor in SSMBR and CMBR at two different stages (within and after 7 days of operation) during the operation period.

Day	Reactor	SMP			
		PN <sup>a</sup> (mg/L)	PS <sup>b</sup> (mg/L)	PN/PS ratio	SMP (mg/L)
Stage I (day 1–7)	SSMBR	9.9–10.2	7.2–9.4	1.1–1.4	7.4–17.4
	CMBR	10.6–10.8	13.5–14.4	0.7–0.8	24.1–25.2
Stage II (after day 7)	SSMBR	1.0–4.4	1.0–6.9	0.3–2.3	1.5–9.2
	CMBR	0.4–5.7	1.0–5.8	0.1–3.2	1.1–9.8

<sup>a</sup> PN, proteins.

<sup>b</sup> PS, polysaccharides.

**Table 3**

Bound EPS compositions and total bound EPS concentrations of mixed liquor in SSMBR and CMBR at two different stages (within and after 7 days of operation) during the operation period.

Day	Reactor	Bound EPS			Total EPS (mg/L)
		PN <sup>a</sup> (mg/L)	PS <sup>b</sup> (mg/L)	PN/PS ratio	
Stage I (day 1–7)	SSMBR	7.4–9.9	9.4–11.8	0.6–1.1	19.2–19.3
	CMBR	9.3–9.9	1.0–9.4	4.7–9.3	10.3–19.3
Stage II (after day 7)	SSMBR	9.8–10.6	1.6–7.5	1.3–6.6	12.2–17.3
	CMBR	6.5–10.1	5.8–14.5	0.7–1.4	12.3–24.6

<sup>a</sup> PN, proteins.

<sup>b</sup> PS, polysaccharides.

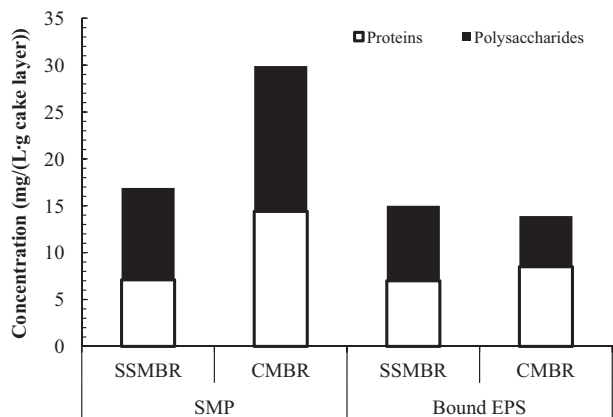


Fig. 2. Compositions of bound EPS and SMP in the cake layer in SSMBR and CMBR.

and  $SMP_p$  on the sponge and biodegradation by attached microorganisms, reduction of cake layer could be also attributed to physical clearance mechanism of sponge, such as frictional force exerted by circulating media on submerged membrane, solute back-transport effect from the membrane surface to the bulk solution due to turbulence of suspended carriers, and membrane shaking by the impact of suspended carriers against them (Lee et al., 2006; Yang et al., 2006).

Since particles could lead to severe membrane fouling by pore blocking and cake formation on the membrane (Lim and Bai, 2003), the CMBR contained smaller sludge flocs and induced higher TMP increment rate (Fig. 1), which illustrated that the presence of smaller sludge flocs contributed to higher  $R_c$  and  $R_p$  in the CMBR. As larger particles could not easily deposit on membrane surface due to higher shear induced diffusion and inertial lift force, SSMBR demonstrated significantly lower membrane fouling propensity (Pan et al., 2010).

In addition, as above-mentioned in Section 3.4, SMP in activated sludge appeared as a major contribution to initial membrane fouling. However, in later stage, membrane fouling development was mainly governed by bound EPS in activated sludge. It has been shown that SMP could increase fouling tendency due to the combined effects of pore clogging and adsorption on membrane walls and within membrane pores (Shen et al., 2012). Thus, higher SMP content of the CMBR cake layer led to higher  $R_p$ , which was well consistent with the results by Jamal Khan et al. (2012). Besides, higher concentration of bound EPS in activated sludge could also increase both  $R_c$  and  $R_p$  in the CMBR. Ng et al. (2006) observed a thick fouling layer on the membrane consisting of microbial cells covered with EPS, which blocked membrane pores. Similar results were also found by Meng et al. (2006b) that the total amount of EPS had a significant positive correlation with the fouling resistance caused by pore blocking and cake formation.

Previous studies have reported that PN/PS ratio in EPS or SMP had a significant impact on filtration resistance as well as fouling propensity (Lee et al., 2003; Tian et al., 2011; Yao et al., 2011). In this study, as both SMP and EPS (especially  $SMP_c$  and  $EPS_c$ ) were responsible for membrane fouling in the CMBR, a new fouling indicator ( $(SMP_c/SMP_p)/(EPS_c/EPS_p)$ ) has been developed. There was a strong correlation between fouling rate and fouling indicator ( $(SMP_c/SMP_p)/(EPS_c/EPS_p) = 9.6727 (dTMP/dt) - 8.3431$ ,  $R^2 = 0.9783$ ). Generally, polysaccharides can penetrate into the cake layer and membrane pores, as well as lead to irreversible fouling due to their partially hydrophilic nature comparing to proteins (Kimura et al., 2004; Meng et al., 2009; Guo et al., 2012). Hence,  $SMP_c$  can be a greater contribution to irreversible fouling than  $EPS_c$ . When activated sludge has higher  $SMP_c$  concentration but lower  $EPS_c$  concentration, the value of  $(SMP_c/SMP_p)/(EPS_c/EPS_p)$  will be higher,

indicating more severe membrane fouling and higher fouling rate ( $\Delta TMP/\Delta t$ ), and vice versa.

#### 4. Conclusions

An in-depth analysis of membrane fouling behaviour in SSMBR and CMBR for synthetic wastewater treatment is presented. SMP and bound EPS of activated sludge in the CMBR governed membrane fouling in the initial stage and later stage, respectively. However, sponge addition could mitigate membrane fouling significantly by preventing pore blocking and reducing cake layer formation. In the SSMBR, lower  $R_c$  and  $R_p$  were ascribed to lower biomass growth, lower sludge viscosity, less filamentous bacteria, larger sludge flocs, as well as lower concentrations of SMP and bound EPS in activated sludge.

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#### Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at <http://dx.doi.org/10.1016/j.biortech.2014.02.111>.

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