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

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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

1 membrane fouling in SSMBR were also attributed to larger particle size, higher zeta
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3 potential and relative hydrophobicity of sludge flocs.
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8 **Keywords:** Submerged membrane bioreactor; Sponge; Attached growth; Membrane
9 fouling; Cake layer
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15 **1. Introduction**

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1 membrane fouling after 70 days of operation as well as less backwash frequency. A
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3 chemical cleaning-in-place (CIP) was investigated by Wei et al. (2011) in a long-term
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5 operation of pilot-scale submerged MBR for municipal wastewater treatment. They
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7 reported that the chemical CIP, in both transmembrane pressure (TMP) controlling
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9 mode and time controlling mode, effectively removed the fouling in terms of membrane
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11 pore blockage and gel layer caused by colloids and soluble organic substances. Wu and
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13 He (2012) suggested that the low irreversible fouling was found in the cyclic aeration
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15 mode, which could be ascribed to the floc destruction and re-flocculation processes.
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17 During the short high aeration period, the preservation of the strong strength bonds
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19 within activated sludge flocs caused less release of soluble and colloidal material in the
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21 supernatant. The weak strength bonds damaged in the high aeration period could be
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23 recovered in the re-flocculation process in the low aeration period.
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33 In addition, using biomass carriers (e.g. plastic media, powdered activated carbon
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35 (PAC), sponge) in MBR is an effective and promising method to control membrane
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37 fouling. Jin et al. (2013) suggested that biomass flocs were less easily broken up with
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39 addition of relatively light and large-sized suspended carriers (AnoxKaldnes, K1
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41 carriers) in ceramic SMBR. Moreover, both extracellular polymer substances (EPS) and
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43 soluble microbial products (SMP) were lower in the SMBR with carriers than those in
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45 the SMBR without carriers. Ng et al. (2013) indicated that higher concentration of fresh
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47 PAC in the SMBR could provide better simultaneous adsorption, decomposition, and
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49 biodegradation effects for the reduction of fouling components in the supernatant of the
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51 mixed liquor such as EPS, fine colloids and planktonic cells. As an idea attached growth
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53 media, sponge has also exhibited excellent performance during biological treatment due
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1 to its advantages of high internal porosity and specific surface area, high stability to
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3 hydrolyses, light weight and low cost (Ngo et al., 2006). When employing in MBRs, it
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5 can act as a mobile carrier for active biomass, reduce cake layer formation on the
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7 membrane surface and retain microorganisms by incorporating both their attached
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9 growth and suspended growth (Ngo et al., 2008). Guo et al. (2008) investigated the
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11 effects of sponge addition on sustainable flux and membrane fouling. They found that
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13 compared to SMBR alone, the suspended sponge cubes in the sponge-submerged
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15 membrane bioreactor (SSMBR) with sponge volume fraction of 10% could significantly
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17 reduce the membrane fouling as well as improve sustainable flux by 2 times. Nguyen et
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19 al. (2012) also confirmed that SSMBR had lower TMP development than that of
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21 conventional SMBR during primary effluent treatment. Meanwhile, SSMBR could
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23 maintain good microbial activity and constant sludge volume index value.
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1 The experiments were conducted using a synthetic wastewater to avoid any
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3 fluctuation in the feed concentration and provide a continuous source of biodegradable
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5 organic pollutants such as glucose, ammonium sulfate and potassium dihydrogen
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7 orthophosphate. It was used to simulate domestic wastewater just after primary
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9 treatment. The synthetic wastewater has dissolved organic carbon (DOC) of 100–130
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11 mg/L, chemical oxygen demand (COD) of 330–360 mg/L, ammonium nitrogen (NH₄-N)
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13 of 12–15 mg/L and orthophosphate (PO₄-P) of 3.3–3.5 mg/L. NaHCO₃ or H₂SO₄ was
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15 used to adjust pH to 7.
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23 *2.2. Experimental setup and operating conditions*

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25 A SSMBR and a CMBR with the same effective working volume were operated in
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27 parallel to compare the performance and membrane fouling behavior. For each MBR, a
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29 polyvinylidene fluoride (PVDF) hollow fiber module with a pore size of 0.2 μm and
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31 surface area of 0.1 m² was used. Both MBRs were filled with sludge from a local
32
33 Wastewater Treatment Plant and acclimatized to synthetic wastewater. They were
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35 started with identical seeding activated sludge with similar initial sludge concentration
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37 (7.03 g/L for SSMBR, 6.98 g/L for CMBR). No sludge was withdrawn from both
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39 MBRs. The reticulated porous polyester-polyurethane sponge (PUS) was used in
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1 using a feeding pump to control the feed rate while the effluent flow rate was controlled
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4 by a suction pump. A pressure gauge was used to measure the TMP and a soaker hose
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images of sludge particles obtained by the Olympus System Microscope Model BX41

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1 These results indicated that sponge addition could significantly mitigate membrane
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3 fouling, which is further discussed in details in Section 3.5.
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6 **Fig. 1.**
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10 *3.2. Mixed liquor suspended solids (MLSS) concentration and apparent viscosity*

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12 During the experimental period, sludge concentration kept increasing in both MBRs
13 due to no sludge withdrawal. MLSS concentrations were 11.50 ± 4.52 g/L and $9.41 \pm$
14 2.38 g/L in the CMBR and SSMBR after 35 and 90 days of operation, respectively. The
15 lower MLSS concentration in the SSMBR might be attributed to the fact that sponge
16 addition could balance the microorganism growth in suspended activated sludge as well
17 as on and inside the porous sponge cubes (Ngo et al., 2006). It was found that there is
18 an exponential relationship between MLSS concentration and sludge viscosity (Reid et
19 al., 2008). In this study, sludge viscosity was higher (3.30 ± 0.50 mPa·s) in the CMBR
20 than that (2.60 ± 0.40 mPa·s) in the SSMBR, demonstrating that higher sludge viscosity
21 was attributed to higher MLSS concentration. In addition, it has been reported that the
22 sludge flocs with excess filamentous bacteria showed high viscosity due to presence of
23 high EPS concentration (Meng et al., 2006a). Overgrowth of filamentous bacteria was
24 found in the CMBR on day 14, whereas there were less filamentous bacteria in the
25 SSMBR until 83 days, which revealed that higher sludge viscosity in the CMBR was
26 also due to abundance of filamentous bacteria. Similar observations were also recorded
27 by Meng et al. (2007) who suggested that sludge viscosity was influenced by MLSS
28 concentration, EPS and filamentous bacteria.
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57 *3.3. Zeta potential, relative hydrophobicity (RH) and particle size distribution*

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1 It has been demonstrated that the flocculation ability of sludge flocs is affected by
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3 their hydrophobicity and surface charge, which positively influences the hydrophobic
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5 interaction and electrostatic repulsion, respectively (Liao et al., 2001; Mikkelsen and
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7 Keiding, 2002). In this study, activated sludge in the SSMBR had higher zeta potential
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9 (- 6.85 ± 3.65 mV) and higher RH (81.00 ± 7.80%) than those in the CMBR (zeta
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11 potential of -10.50 ± 4.50 mV, RH of 63.13 ± 13.60%). The results indicated that there
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13 might be a positive relationship between surface charge (zeta potential) and
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15 hydrophobicity of activated sludge. Additionally, Meng et al. (2006a) reported that
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17 excess filamentous bacteria could prevent the agglomeration of floc particles by
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19 producing a bridge lattice due to the generation of abundant filaments from the flocs
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1 similar bound EPS concentrations, slightly higher protein concentrations (EPS_P) but
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3 significantly lower polysaccharide concentrations (EPS_C) were obtained in the CMBR.
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5 After 7 days of operation, the SMP concentrations (including SMP_P and SMP_C) of both
6
7 MBRs presented minor difference. On the other hand, bound EPS concentrations
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9 (12.3–24.6 mg/L) in the CMBR were higher than those in the SSMBR (12.2–17.3
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11 mg/L), with lower protein concentrations (EPS_P) but significantly higher polysaccharide
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13 concentrations (EPS_C). In this study, increase of sludge concentration under infinite
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15 SRT condition induced the decrease in food to microorganism (F/M) ratio ($0.1-0.2\text{ d}^{-1}$).
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17 As a consequence, both MBRs were fed with limited available substrate, which could
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19 cause more cell lysis and cell hydrolysis, thereby releasing EPS and SMP in activated
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21 sludge (Yigit et al., 2008). Moreover, the excess growth of filamentous bacteria could
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23 produce more SMP, resulting in severe fouling (Pan et al., 2010). Therefore, the CMBR
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25 exhibited more serious fouling compared with the SSMBR. In the SSMBR, it was
26
27 obvious that sponge addition could reduce SMP_C concentrations during the first 7-day
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29 run and EPS_C afterwards by the means of adsorption onto sponge as well as
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31 biodegradation by attached biomass of the sponge.
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43 It has been reported that large quantity of EPS in activated sludge increased floc
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45 strength by polymer entanglement, thereby increasing the extent of sludge flocs
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47 agglomeration (Mikkelsen and Keiding, 2002). However, in this study, lower EPS
48
49 concentration but larger particles were observed in the SSMBR, pointing out that the
50
51 flocculation ability of sludge flocs may not only depend on EPS concentration. Lee et al.
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53 (2003) found that the ratio of proteins to polysaccharides (PN/PS ratio) in EPS was
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55 important in controlling the hydrophobicity and surface charge of sludge flocs. Table 3
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1 shows that a significantly higher PN/PS ratio in bound EPS was found in the SSMBR
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3 after 7 days operation. Higher RH of activated sludge in the SSMBR proved that higher
4
5 EPS_P concentration increased the hydrophobicity of sludge flocs by providing amino
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7 acids with more hydrophobic side groups, while lower EPS_C concentration contributed
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9 to less hydrophilic nature of sludge. Moreover, the amino groups in EPS_P containing
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11 positive charges neutralized some of negatively charged activated sludge, thereby
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13 inducing higher zeta potential of sludge flocs in the SSMBR (Lee et al., 2003; Liao et
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15 al., 2001). Thus, PN/PS ratio in bound EPS could positively influence hydrophobicity
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17 and zeta potential of activated sludge, thereby having an impact on the agglomeration
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19 ability of the flocs.
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25 **Table 2.**

26 **Table 3.**

32 *3.5. Membrane fouling behaviour*

34 Results of fouling resistance showed that the CMBR had a higher total resistance
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36 (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
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38 resistance (R_M) were the same ($1.71 \times 10^{12} \text{ m}^{-1}$) for both MBRs. Higher cake layer
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40 resistance (R_C) was found for the CMBR than that for the SSMBR, corresponding to
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42 $3.04 \times 10^{12} \text{ m}^{-1}$ and $0.85 \times 10^{12} \text{ m}^{-1}$, respectively. Moreover, pore blocking resistance
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44 (R_P) for the CMBR was notably higher. R_P of the CMBR accounted for about 20% of
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46 R_T , whereas there was no R_P in the SSMBR. These results suggested that cake layer
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48 formation was one of the main factors contributing to membrane fouling. Furthermore,
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50 sponge could alleviate membrane fouling not only by preventing pore blocking but also
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52 by reducing cake layer formation. Some researchers (Jamal Khan et al., 2012; Yang et
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1 al., 2006) have reported the similar findings that R_C was major fraction of R_T and
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3 sponge addition could reduce R_C .
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8 As discussed in Section 3.2, activated sludge in both MBRs possessed different
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10 properties, which were correlated with membrane fouling potential as well as fouling
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12 resistance. Higher MLSS concentration could lead to formation of a sticky cake layer on
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14 membrane surface due to higher sludge viscosity (Itonaga et al., 2004). Additionally,
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16 the sludge flocs with abundance of filamentous bacteria would more easily deposit on
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18 membrane surface due to its high viscosity, causing the formation of a non-porous cake
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20 layer (Meng et al., 2006a). Therefore, it could be noted that higher MLSS concentration
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22 and overgrowth of filamentous bacteria contributed to formation of sticky and non-
23
24 porous cake layer, giving rise to higher R_C in the CMBR. Being the major fraction of
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26 the total fouling resistance, the cake layer was analysed with respect to EPS and SMP
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28 (including polysaccharides and proteins). Fig. 2 shows the composition of EPS and
29
30 SMP in the cake layer on membrane surface for both SSMBR and CMBR. Bound EPS
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32 concentrations were similar for the SSMBR (15.0 mg/(L·g cake layer)) and the CMBR
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34 (13.9 mg/(L·g cake layer)). However, higher concentrations of SMP_C and SMP_P (14.4
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36 and 15.5 mg/(L·g cake layer), respectively) were obtained for the CMBR, while SMP_C
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38 and SMP_P of the cake layer were comparatively lower for the SSMBR (9.8 and 7.1
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40 mg/(L·g cake layer), respectively). These results elucidated that higher R_C in the CMBR
41
42 was mainly caused by SMP (including SMP_C and SMP_P) on membrane surface. At high
43
44 TMP, more SMP_C and SMP_P could be adsorbed and/or attached onto membrane surface
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46 due to the high drag force provided by permeate pump. On contrary, sponge addition
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48 effectively reduced SMP_C and SMP_P in cake layer on membrane surface. Apart from
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1 adsorption of SMP_C and SMP_P on the sponge and biodegradation by attached
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3 microorganisms, reduction of cake layer could be also attributed to physical clearance
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5 mechanism of sponge, such as frictional force exerted by circulating media on
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7 submerged membrane, solute back-transport effect from the membrane surface to the
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9 bulk solution due to turbulence of suspended carriers, and membrane shaking by the
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11 impact of suspended carriers against them (Lee et al., 2006; Yang et al., 2006).
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15 **Fig. 2.**

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20 Since particles could lead to severe membrane fouling by pore blocking and cake
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22 formation on the membrane (Lim and Bai, 2003), the CMBR contained smaller sludge
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24 flocs and induced higher TMP increment rate (Fig. 1), which illustrated that the
25
26 presence of smaller sludge flocs contributed to higher R_C and R_P in the CMBR. As
27
28 larger particles could not easily deposit on membrane surface due to higher shear
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30 induced diffusion and inertial lift force, SSMBR demonstrated significantly lower
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32 membrane fouling propensity (Pan et al., 2010).
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40 In addition, as above-mentioned in Section 3.4, SMP in activated sludge appeared
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42 as a major contribution to initial membrane fouling. However, in later stage, membrane
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44 fouling development was mainly governed by bound EPS in activated sludge. It has
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46 been showed that SMP could increase fouling tendency due to the combined effects of
47
48 pore clogging and adsorption on membrane walls and within membrane pores (Shen et
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50 al., 2012). Thus, higher SMP content of the CMBR cake layer led to higher R_P , which
51
52 was well consistent with the results by Jamal Khan et al. (2012). Besides, higher
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54 concentration of bound EPS in activated sludge could also increase both R_C and R_P in
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1 the CMBR. Ng et al. (2006) observed a thick fouling layer on the membrane consisting
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3 of microbial cells covered with EPS, which blocked membrane pores. Similar results
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5 were also found by Meng et al. (2006b) that the total amount of EPS had a significant
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7 positive correlation with the fouling resistance caused by pore blocking and cake
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9 formation.
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15 Previous studies have reported that PN/PS ratio in EPS or SMP had a significant
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17 impact on filtration resistance as well as fouling propensity (Lee et al., 2003; Tian et al.,
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19 2011; Yao et al., 2011). In this study, as both SMP and EPS (especially SMP_C and EPS_C)
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21 were responsible for membrane fouling in the CMBR, a new fouling indicator
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1 the CMBR governed membrane fouling in the initial stage and later stage, respectively.
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3 However, sponge addition could mitigate membrane fouling significantly by preventing
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5 pore blocking and reducing cake layer formation. In the SSMBR, lower R_C and R_P were
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7 ascribed to lower biomass growth, lower sludge viscosity, less filamentous bacteria,
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9 larger sludge flocs, as well as lower concentrations of SMP and bound EPS in activated
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Table Titles

Table 1 Removal efficiency of DOC, COD, PO₄-P, NH₄-N and TN in SSMBR and CMBR during the operation period.

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.

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.

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Table 1

Removal efficiency of DOC, COD, PO₄-P, NH₄-N and TN in SSMBR and CMBR during the operation period.

Reactors	DOC (%)	COD (%)	PO ₄ -P (%)	NH ₄ -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

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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 ^a (mg/L)	PS ^b (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

^a PN, proteins; ^b PS, polysaccharides

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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			
		PN ^a (mg/L)	PS ^b (mg/L)	PN/PS ratio	Total EPS (mg/L)
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

^a PN, proteins; ^b PS, polysaccharides

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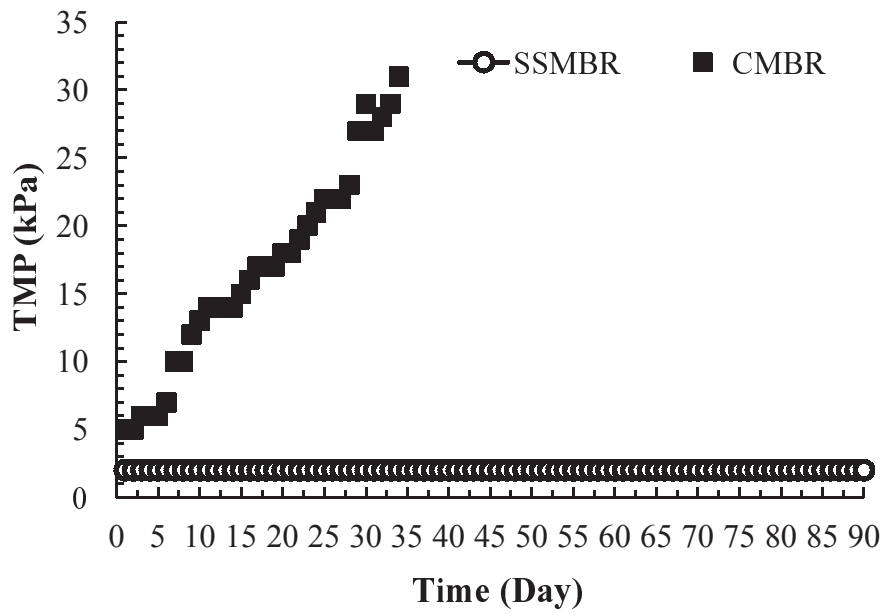


Fig. 1.

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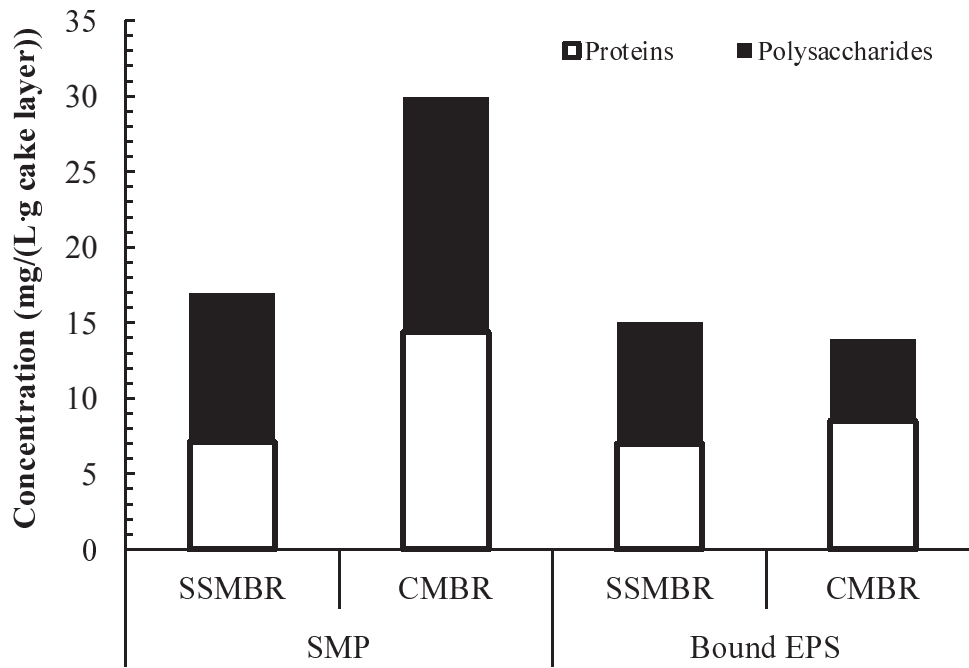


Fig. 2.

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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.