Performance and membrane fouling of two types of laboratory-scale submerged membrane bioreactors for hospital wastewater treatment at low flux condition

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Abstract
The performance and membrane fouling of a lab-scale submerged sponge-membrane bioreactor (Sponge-MBR) and a conventional MBR were investigated and compared for hospital wastewater treat- ment at low fluxes of 2–6 LMH. COD removal by the Sponge-MBR was similar to that of the MBR, while the Sponge-MBR achieved 9–16% removed more total nitrogen than the MBR. This was due to 60% of total biomass being entrapped in the sponges, which enhanced simultaneous nitrification denitrification. Additionally, the fouling rates of the Sponge-MBR were 11-, 6.2- and 3.8-times less than those of the MBR at flux rates of 2, 4 and 6 LMH, respectively. It indicates the addition of sponge media into a MBR could effectively reduce the fouling caused by cake formation and absorption of soluble substances in a low flux scenario.

Keywords: Hospital wastewater; Fouling; Sponge membrane bioreactor; Low flux

1. Introduction

Membrane Bioreactor (MBR) has several advantages compared to conventional activated sludge (CAS), namely higher quality effluent, less area requirement, higher biomass concentration and less sludge production [1,2]. However, membrane fouling is a major obstacle to the widespread application of MBRs, and it causes declining permeate flux, increases operational costs and shortens membrane life [3,4]. The factors affecting membrane fouling can be divided into three overarching types, specifically membrane characteristics, biomass and operating conditions [5]. Many studies have been conducted on reducing membrane fouling, e.g. enhancing sludge retention time [6,7], operating MBRs at low flux [8,9], applying air sparing and back flushing [10], modifying sludge properties by adding flocculant or adsorbent [11–16], etc. Of these methods the one attracting much attention is a hybrid membrane bioreactor (HMBR) using suspended carriers as supporting media for biofilm development in the membrane tank. Adding sponges or fluidized media into MBR can reduce membrane fouling by enhancing the combination of suspended and colloidal particles on the medium's surface, and reduce clogging on the membrane surface by the collision between moving medium and membrane surface [3,9,17]. Furthermore, adding sponges also improves the efficiency of biodegradation and enhances the nitrification process [3].

To treat synthetic wastewater, Khan et al. [18] compared the performance of a MBR and a Sponge-MBR (sponge volume occupied 15% reactor volume). Results indicated that the Sponge-MBR effectively removed TN and TP (89% and 58%, respectively), compared to the MBR (74% and 38%, respectively). Liu et al. [3] showed that the speed of TMP increment in the Sponge-MBR was apparently slowed down. When the TMP reached 20 kPa, the Sponge-MBR operated for more than 92 days while the MBR operated for only 57–65 days. Their study also reported that in the Sponge-MBR, the average removals of COD, NH_4^+-N, TN and TP were improved by 3.8%, 4.2%, 13.7% and 1.7%, respectively. Yang et al. [19] conducted a hybrid MBR with porous, flexible suspended carriers to treat terephthalic acid wastewater. The MBR was efficient in controlling membrane fouling, especially the cake layer on the membrane, with 86% reduction in cake resistance and 20% of critical flux increase compared to the MBR.

Hospital wastewater contains harmful pollutants such as pathogenic microorganisms (bacteria and viruses), heavy metal (Pb), biodegradable organic material (protein, fat, carbohydrates)

[20,21] and pharmaceuticals such as antibiotics, endocrine disrupting compounds (EDCs), residue chemicals (phenol, chloroforms) [22–24]. Kovalova et al. [23] reported that a pilot-scale MBR could eliminate approximately 60% of major antibiotics but only 22% of all measured pharmaceuticals and metabolites in Swiss hospital wastewater. The pollutants from hospital wastewater can easily reach water bodies and cause aquatic pollution and human health problems. Thus, the treatment of hospital wastewater is critical in order to reduce damage to the environment and protect human health. In addition, due to the actual situation of hospitals such as their limited space and population, MBR technology has emerged as the most suitable technology for hospital wastewater treatment. The Sponge-MBR is advantageous in terms of functioning as an anti-fouling solution and removing pollutants [1]. While operating at low flux, MBR can treat wastewater containing high strength concentrations [9] or pharmaceuticals [24]. The low flux MBR coupled with sponge media could create the conditions in which microbial biodiversity could thrive, and long attached biomass retention. This could effectively treat the hospital wastewater and consequently, the study aims to compare the treatment performance and fouling characteristics of MBR and Sponge-MBR treated hospital wastewater at low flux conditions.

2. Material and methods

2.1. MBRs and operating conditions

Two lab-scale submerged MBRs operated in parallel each with a working volume of 22 L (L × W × H = 0.28 m × 0.14 m × 0.55 m). Each PVDF hollow-fiber membrane module (Motimo, China) had a surface area of 0.5 m^2 and pore size of 0.2 lm. The systems were controlled automatically by timers, solenoid valves and digital pressure gauges. Air diffusers were placed at the bottom of the reactor and at the rear end of the membrane module for aeration and air scouring. Dissolved oxygen was maintained at higher than 4 mg/L by the air blowers with the air supply of 70 L/m^3 min. The MBRs' permeate pumps were operated in a cyclic mode (8 min on/2 min off). For each permeate flux, the membrane was externally cleaned by chemicals (0.5% NaOCl) for 4 h. The digital pressure gauges recorded the trans-membrane pressure (TMP) daily. A schematic illustration of the MBR systems is presented in Fig. 1.

The seed activated sludge was collected from a full-scale MBR in Ho Chi Minh City, Vietnam. The amount of the initial mixed liquor suspended solids (MLSS) was approximately 5000 mg/L. The sludge retention time (SRT) was maintained at 45 days during operation. The operating conditions of the MBRs are presented in Table 1.

The Sponge-MBR used polyethylene cubic sponges with a porosity of 98% and dimensions of 2 cm × 2 cm × 2 cm. Initially, the sponges were added in one MBR with the amount of 20% serving as the reactor volume.

2.2. Hospital wastewater

Wastewater was directly collected daily from the equalization tank of the wastewater treatment plant of a hospital in Ho Chi Minh City. The hospital is nearby university with 900 beds and 1100 staffs. The influent wastewater then was stored in a 60-L tank to feed into the MBRs. The composition of wastewater is presented in Table 2.

2.3. Analytical methods

Parameters of COD, TKN, NH_4^+-N, NO_2-N, NO_3-N, TN, TP, MLSS and MLVSS were determined according to standard methods [25]. The biomass attached in sponges was converted into MLSS concentration. Twice a week, five (5) sponges were collected randomly to analyze sponge MLSS. Sludge in five (5) sponges was taken out by carefully squeezing solids into a certain volume of distilled water to obtain squeezed solution significantly. Duplicate sampling for sponge MLSS measurement was applied. Monthly the number of new sponges were added to compensate the loss through sampling. The MLSS in sponges was calculated based on the number of sponges in MBR and suspended solids concentration in squeezed solution.

A specific fraction of the MBR supernatant was achieved by centrifuging: the mixed liquor sludge sample at 4000 rpm and 4 oC for 10 min (Universal 320, Hettich, Germany). TMP was recorded daily

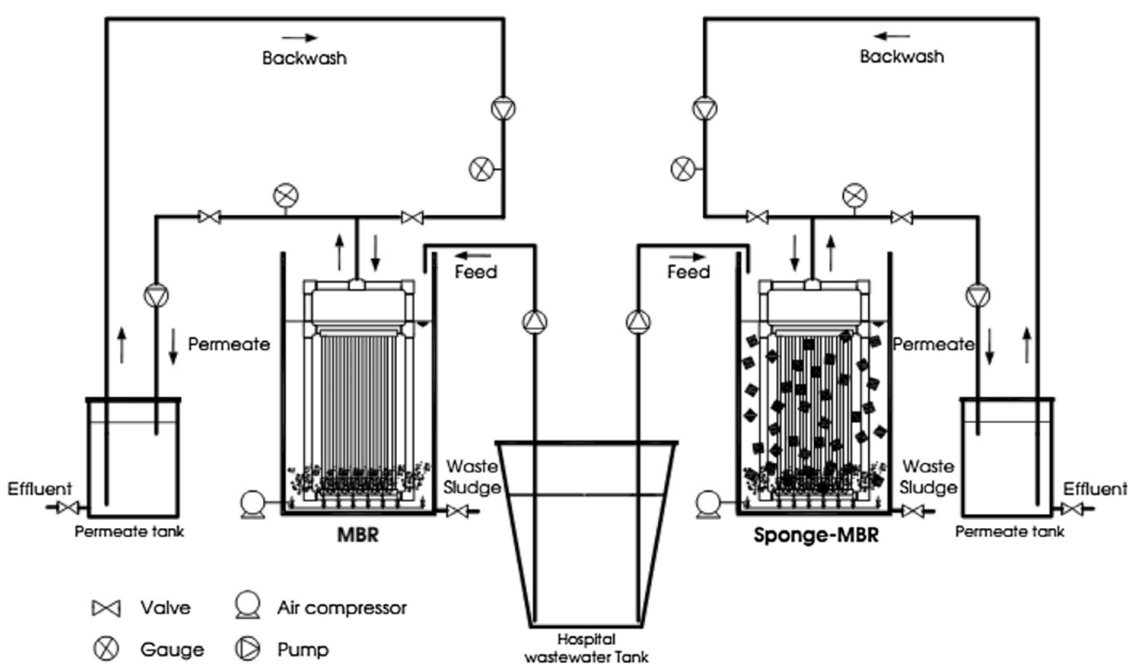

Fig. 1. Schematic illustration of the MBR systems.

Table 1
Operating conditions of MBRs.

Operating parameters	2 LMH		4 LMH		6 LMH	
	Sponge-MBR	MBR	Sponge-MBR	MBR	Sponge-MBR	MBR
F/M (kg COD/kg MLSS day)	0.047 ± 0.01	0.054 ± 0.01	0.072 ± 0.01	0.074 ± 0.02	0.106 ± 0.03	0.123 ± 0.04
OLR (kg COD/m^3 day)	0.15 ± 0.04		0.23 ± 0.07		0.39 ± 0.13	
HRT (h)	22.0		11.0		7.3	

Table 2
Composition of used hospital wastewater.

Parameters	Unit	Average value (max – min)
Temperature	oC	29 (28–30)
pH	–	6.8–8.2
COD	mg/L	123 (38–224)
TSS	mg/L	75.1 (26.8–124.6)
NH_4^+-N	mg/L	23.3 (9–38.4)
NO_3-N	mg/L	<0.1
TKN	mg/L	32.3 (19.6–57.1)
TP	mg/L	3.3 (1.3–5.5)

and fouling rate (dTMP/dt) was determined by estimating the slope between TMP over time at the linear segment.

Nitrogen balance was calculated using Eq. (1). Nitrogen assimilated into the biomass was estimated based on the assimilated nitrogen of 12% VSS [26]. Balancing the nitrogen helped to evaluate the simultaneous nitrification denitrification (SND) that occurred in the Sponge-MBR:

$$TN_{in} = TN_{out} + TN_{assimilated} + TN_{denitrification} \quad (1)$$

According to the resistance-in-series model, the resistance of MBRs can be calculated employing the Darcy equation (Eqs. (2) and (3)):

$$J = \frac{\Delta P}{\mu \cdot R_t} \quad (2)$$

$$R_t = R_m + R_c + R_f \quad (3)$$

where J is the permeate flux; DP is trans-membrane pressure (TMP); l is the viscosity of permeate; R_t is the total resistance; R_m is the intrinsic membrane resistance; R_c is the cake resistance and R_f is the fouling resistance which caused by the adsorption of soluble matters and pore blocking.

Flux (J) and TMP data were used to calculate the component resistances based on Eqs. (2) and (3). Total resistance (R_t) was calculated from the final flux and TMP values when the operation ended by the filtration of pure water using the membrane. The cake resistance (R_c) related to deposition of the cake layer on membrane surface that can be washed out manually under tap water. Thus, the total of (R_f + R_m) can be obtained by the filtration of pure water with the membrane after removing the cake layer. R_c can be evaluated as subtraction of the total resistance (R_t) and the total of (R_f + R_m). This membrane, then, was chemically cleaned by soaking it for 4 h in a solution of 0.5% NaOCl and NaOH 4% to determine the lasting resistance membrane (R_m) by the filtration of pure water. Finally, the R_f is determined by subtracting this for R_m.

3. Results and discussions

3.1. COD removal

The average COD concentration and removal efficiency during the operating periods are shown in Fig. 2. Regardless of the variation in raw wastewater (COD = 96–224 mg/L), the average COD values in the membrane permeate were as low as 11–16 mg/L at a flux of 2–6 LMH. The permeate COD concentration of both MBRs reached class A of the Vietnam National Technical Regulation on health care wastewater - QCVN 28: 2010/BTNMT (50 mgCOD/L). The average COD removal rate of Sponge-MBR and MBR was about 0.04–0.10 mgCOD/mgMLVSS h at fluxes of 2–6 LMH.

A similar observation was reported by Wen et al. [27], who suggested the COD in the permeate of a MBR treating hospital wastewater was always less than 30 mg/L with an 80% COD removal efficiency.

The average COD removal efficiencies of Sponge-MBR were 89 ± 9%, 88 ± 6% and 85 ± 10% for the fluxes of 2, 4 and 6 LMH, respectively, while those concerning the MBR were 84 ± 10%, 86 ± 6% and 84 ± 10%. This result indicates that the Sponge-MBR's removal of COD was quite similar to that of the MBR at the low flux range with a F/M ratio of 0.05–0.12 day [1]. A similar outcome was reported by Liu et al. [3], who showed that COD removal improved by only 3.8% in Sponge-MBR compared with the MBR.

3.2. Nitrogen removal

The average concentrations of TKN, NH_4-N, NO_2-N, NO_3-N and TN in membrane permeates are summarized in Table 3. The effluent concentrations of NH_4^+-N and NO_3-N in both MBRs meet the requirements of the Vietnam National Technical Regulation on health care wastewater - QCVN 28:2010/BTNMT (10 mg NH_4^+-N/L and 30 mg NO_3-N/L). In addition, the average NO_2-N in permeates of both MBRs were approximately 0.2 mg/L. During the operation period, average ammonia removal efficiencies of 100%, 99% and 99% were observed in both MBRs at fluxes of 2, 4 and 6 LMH, respectively, with the operating HRTs in the 7.3–22 h range. These results were in line with those recorded by Liu et al. [3]. There was no significant improvement between HRTs of 4 h and 8 h in ammonia removal efficiencies, as almost all (99%) had been removed after 4 h. Results show that like domestic wastewater treatment, both MBRs could achieve high levels of nitrification when treating hospital wastewater. Gender et al. [28] stated that the nitrification capacity of the MBR is greater than the conventional activated sludge due to higher sludge retention time (SRT). The smaller floc size in the high sludge age MBR helps microorganisms be exposed to oxygen and nutrients much more easily.

The TN removal efficiencies of Sponge-MBR were 52 ± 13%, 36 ± 1% and 25 ± 1% at fluxes of 2, 4 and 6 LMH, respectively, while those of MBR were 36 ± 7%, 27 ± 5% and 12 ± 1% for the operated flux range. The average removal efficiencies of TN in the Sponge-MBR were 9–16% higher than those in the MBR. This was due to the effect of sponge media which can create a simultaneous nitrification and denitrification (SND) state for complete nitrogen removal [29]. Fig. 3 illustrates that the average TN amounts removed due to SND in the Sponge-MBR were 34%, 25% and 15% at fluxes of 2, 4 and 6 LMH. Conversely they were only 13%, 6% and 0.3% in the MBR.

In the sponges, nitrification probably takes place on their surface, whereas anaerobic/anoxic conditions inside the sponge provide a suitable environment for denitrification [30]. A higher HRT enriches slow growing microorganisms and creates effective contacts between microorganisms and substrates. SND occurs in the sponge medium because of the biomass captured within the pores

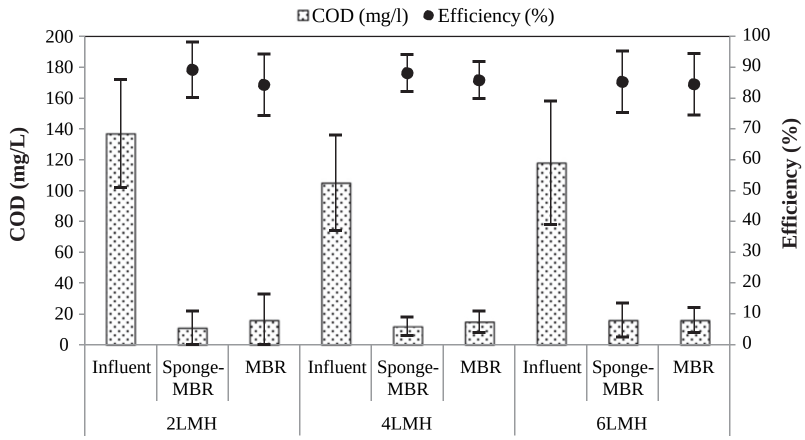

Fig. 2. COD removal in the Sponge-MBR and MBR at various flux.

Table 3
Nitrogen species (mg/L) in the membrane permeates during operation period.

Flux	2 LMH		4 LMH		6 LMH	
Reactor	Sponge-MBR	MBR	Sponge-MBR	MBR	Sponge-MBR	MBR
NH_4^+-N	0.5 ± 1.0	0.4 ± 1.0	0.3 ± 1.0	0.4 ± 1.0	0.2 ± 1.0	1.0 ± 1.0
NO_3^--N	16.7 ± 6.0	18.8 ± 6.0	16.5 ± 9.1	19.0 ± 10.0	21.5 ± 6.0	26.0 ± 6.1
NO_2^--N	0.2 ± 0.1	0.1 ± 0.1	0.3 ± 0.1	0.1 ± 0.1	0.3 ± 0.2	0.2 ± 0.2
TN	18.5 ± 7.0	20.8 ± 8.1	21.9 ± 7.2	24.5 ± 8.3	23.3 ± 5.0	28.7 ± 6.1

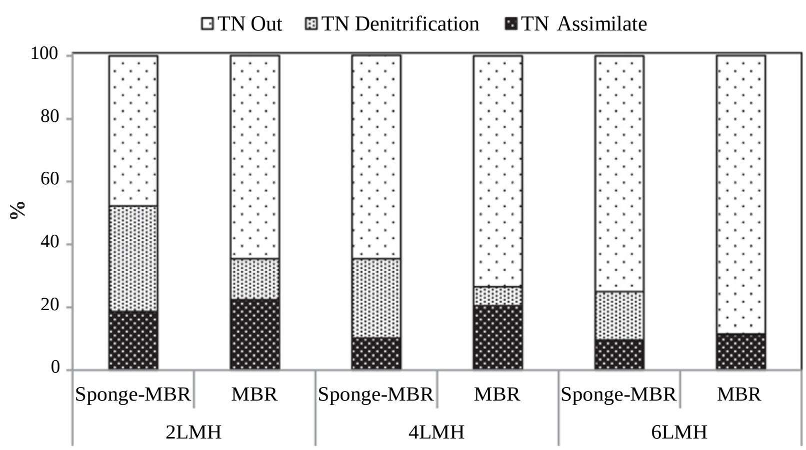

Fig. 3. Nitrogen balance in Sponge-MBR and MBR.

of the sponge and a limited oxygen concentration inside the pores [19]. Thanh et al. [31] compared between sponge MBR and MBR treating catfish pond wastewater. The Sponge-MBR had twice the TN removal capacity at the same 2, 4 and 8 h HRT compared to the MBR. However, the low generated biomass due to low F/M ratio also led to low TP elimination. The TP removal efficiencies in the Sponge-MBR were 28 ± 12%, 22 ± 10% and 26 ± 11% for the fluxes of 2, 4 and 6 LMH, respectively, while those in the MBR were 29 ± 16%, 26 ± 11% and 20 ± 15%.

3.3. Biomass characteristics

Biomass fraction between attached and suspended microorganisms impacted on MBR performance and sludge microbiology. Fig. 4 shows the MLSS concentrations of MBRs and the ratio of sponge MLSS over total MLSS in Sponge-MBR during the operation's duration. The biomass concentration fluctuated between 4889 and 6978 mg/L in the Sponge-MBR and between 3720 and 5825 mg/L in the MBR. At fluxes of 2 and 6 LMH, the MLSS concentrations of Sponge-MBR were higher than those of the MBR. The MLSS concentrations at a flux of 4 LMH were the same in both MBRs due to the Sponge-MBR's influent pump having broken down for days during this period (day 93, 94, 102, 114, and 135). In general, the Sponge-MBR demonstrated superior biomass retention compared to the MBR. This was due to a large amount of biomass attached in the sponges. At the flux of 2 LMH, the sludge concentration of both MBR fluctuated due to the oscillation of influent COD concentration. Additionally, the low operated F/M ratio of the MBRs led to the produced biomass not being able to compensate for the excess biomass.

The average MLVSS/MLSS ratio of the Sponge-MBR was 0.6 and similar to the MBR. However, the average ratio of 0.64 (0.52–0.79) of the sponge-attached biomass was higher than that of suspended biomass of 0.56 (0.49–0.63).

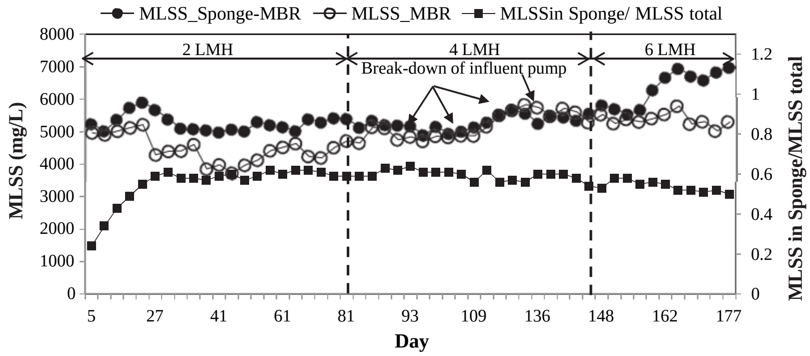

Fig. 4. MLSS concentration and biomass fraction in MBRs.

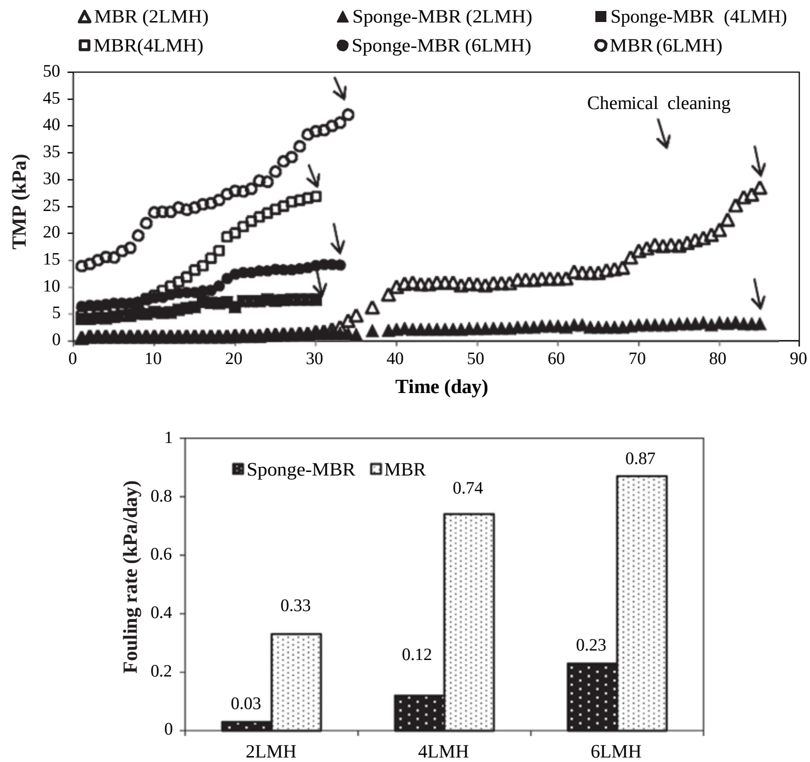

Fig. 5. Evolution of trans-membrane pressure (TMP) (above) and fouling rates of MBRs (below).

Fig. 4 shows that the MLSS in the sponges increased in the first 30 days of operation; the ratio of $MLSS_{sponge}/MLSS_{total}$ also increased during this period and reached about 0.6. Then this ratio stabilized in the next operation period. This means 20% sponge of reactor volume contained up to 60% of total biomass in the Sponge-MBR. In the last 15 days at 6 LMH flux, the ratio tended to decline from 0.55 to 0.50 due to MLSS in the sponges being saturated in the 17,000–18,000 mg/L range. From this period, the suspended biomass increased continuously. In this study, yield coefficient of 0.64 mgVSS/mgCOD in the Sponge-MBR was higher than that of 0.41 mgVSS/mgCOD in the MBR. This means that the attached biomass in the sponges seemed to be more active than the suspended biomass.

3.4. Membrane fouling

Fig. 5 demonstrates TMP development of the MBRs during the period of operation. There was a significant difference in TMP fluctuations between the two MBRs. The TMP of the Sponge-MBR increased from 0.6 to 3.2 kPa in 85 days (2 LMH); 4.1 to 7.7 kPa in 30 days (4 LMH); 6.4 to 14.4 kPa in 35 days (6 LMH), respectively. Nevertheless, the TMP of the MBR developed from 0.6 to

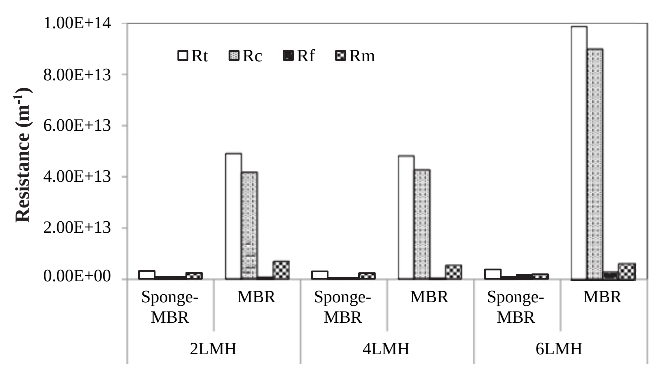

Fig. 6. Membrane resistances.

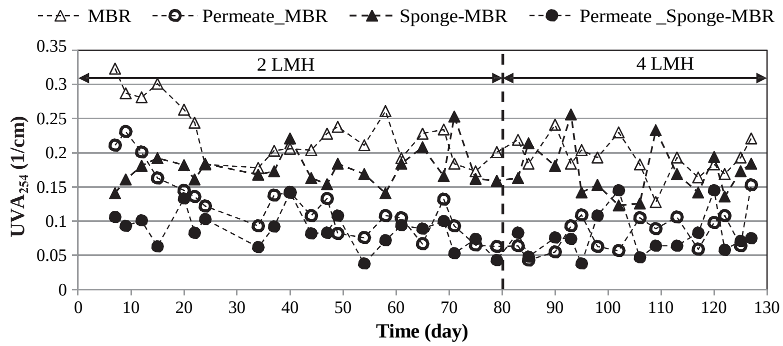

Fig. 7. UVA_{254} value in membrane supernatant and permeate of MBRs during operation.

28.6 kPa in 85 days (2 LMH); 4.8 to 26.9 kPa in 30 days (4 LMH); and 13.9 to 44.2 kPa in 36 days (6 LMH).

The fouling rates of Sponge-MBR were observed to be lower than those of the MBR throughout the operation. At the fluxes of 2, 4 and 6 LMH, the fouling rates of the Sponge-MBR were 0.03, 0.12, and 0.23 kPa/day whereas the fouling rates of the MBR were 0.33, 0.74, 0.87 kPa/day, respectively. Hence, the fouling rates of the MBR were 11, 6.2 and 3.8 times higher at fluxes of 2, 4 and 6 LMH. These results indicated that the Sponge-MBR could reduce membrane fouling efficiently. Other studies have documented similar results [3,6,19] asserted that to reach TMP of 20 kPa the MBR had to operate for 57–65 days whereas the Sponge-MBR functioned for more than 92 days.

Fig. 6 depicts the membrane resistance after the operation has ceased. The results demonstrated that the total membrane resistance (R_t) values of the Sponge-MBR were much lower than that of the MBR. At the end of each operated flux, R_t of Sponge-MBR at fluxes of 2, 4 and 6 LMH were 3.07×10^{12} (1/m), 2.87×10^{12} (1/m), and 3.62×10^{12} (1/m), respectively. Those of the MBR were 4.93×10^{13} (1/m), 4.85×10^{13} (1/m) and 9.95×10^{13} (1/m). It was found that the major resistance component in the Sponge-MBR was the resistance of intrinsic membrane (R_m) while the main resistance component of the MBR was cake resistance (R_c). At the fluxes of 2, 4 and 6 LMH, the cake resistances of the Sponge-MBR were 14%, 13% and 18% of total resistance whereas those of the MBR were 85%, 89% and 91% of total resistance. Thus, the addition of sponge media in the MBR could solve the problem of cake fouling efficiently compared to the MBR. The results also demonstrated a collision between the moving sponges and membrane fibers could, firstly, enhance friction and secondly, reduce the formation of biofilm on the surface of the membrane fibers. Similarly, Yang et al. [19] reported that Sponge-MBR was efficient in controlling membrane fouling, especially the cake layer on the membrane. The result was 86% reduction in cake resistance and an increase by 20% of the critical flux compared to the MBR.

In addition, ultra violet absorbance (UVA_{254}) of membrane supernatant and permeate of both MBRs were measured to confirm that irreversible fouling occurred due to absorption of soluble matters into the membrane. The UVA_{254} value indicated the presence of double bond linkage substances such as protein, humic acid and fulvic acid. Fig. 7 shows that the average UVA_{254} values in supernatant and permeate of the MBR (0.214 ± 0.041 cm ^{1} and 0.108 ± 0.045 cm ^{1}) were higher than those of the Sponge-MBR (± 0.031 cm ^{1} and 0.083 ± 0.028 cm ^{1}). This result reveals that the absorbance values of the membrane permeate or membrane supernatant in MBR were higher than those in the Sponge-MBR. It also confirms that the sponges could help eliminate soluble organic matters effectively. Furthermore, the UVA_{254} values in the membrane permeate were always lower than those in both MBRs' membrane supernatant. This indicated that the soluble organic matters were trapped in the membrane and could then cause irreversible fouling. As a consequence, the sponges can reduce fouling by preventing cake formation and absorption of soluble substances on to the membrane.

4. Conclusions

Based on this study's results, some important assertions can be made as follows:

- Adding sponges into MBR (20% volume) could enhance TN removal by 9–16% at fluxes as low as 2–6 LMH.
- The movement of sponges caused friction force to membrane surface during operation, preventing cake formation on the membrane and reducing cake resistance, and therefore control fouling. The Sponge-MBR's cake resistance was only 13–18% of the total resistance while that of the MBR represented 85–91% under the low flux range.
- Soluble substances being absorbed into the membrane contributed to irreversible fouling for both the Sponge-MBR and MBR. It was observed that the Sponge-MBR generated less soluble matters in both supernatant and permeate compared to MBR.

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