

Effect of hydraulic retention time and temperature on submerged membrane bioreactor (SMBR) performance

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Abstract—Water shortages and strict environmental provisions necessitate wastewater renovation using various wastewater treatment methods, among which applications of submerged membrane bioreactors (SMBRs) are rapidly increasing due to their advantages such as high loading capacity and quality of effluent. In this work, the effect of hydraulic retention time (HRT 8, 10 and 12 h) and temperature (25, 30 and 35 °C) on membrane fouling and sludge production was investigated in a 5-Liter SMBR equipped with immersed PVDF hollow fiber membrane module. Phenolic synthetic wastewater and acclimatized activated sludge with phenol during a 2-month period were used as toxic and microbial sources, respectively. Results showed that by increasing HRT membrane fouling decreases, while excellent treatment performance of over 99.5% phenol and 95% COD removals was achieved at all HRTs. Therefore, HRT=8 h corresponding to the highest effluent flow rate of 12 L/m²·h was used to investigate the effect of temperature, resulting in phenol and COD removals of higher than 99 and 96%, respectively, at all temperatures. Membrane fouling occurred at 12, 5 and 3 days for 25, 30 and 35 °C, respectively. Additionally, the effect of HRT and temperature on mixed liquor volatile suspended solid (MLVSS) as a measure of biomass was examined. MLVSS concentration showed decreases with increasing HRT and temperature. Overall, it was shown that SMBR can be used to efficiently treat phenolic wastewater at a range of flow rates and temperatures, among which HRT=8 h and T=25 °C are the preferred operating conditions, resulting in high flow rate and low membrane fouling.

Key words: Submerged Membrane Bioreactor (SMBR), Hollow Fiber Membrane, Hydraulic Retention Time, Temperature, Membrane Fouling

INTRODUCTION

Water is a national asset. Water shortages and strict environmental provisions create the demand to reuse water. Various treatment methods have been used for wastewater renovation, among which biological treatments such as activated sludge system, biofiltration and moving bed bioreactor (MBBR) have been widely exploited for industrial and municipal wastewater treatment. In the past two decades, membrane bioreactors, specially submerged bioreactors, have attracted the attention of many researchers due to their high loading capacity and effluent quality [1]. SMBRs comprise microfiltration (MF) or ultrafiltration (UF) membrane processes with activated sludge, and are now widely used for municipal and industrial wastewater treatment [1-3]. A variety of membranes are being used in SMBRs such as flat sheet, hollow fiber and tubular membranes [Ref]. However, hollow fiber membranes are distinguished as effective due to their modest energy requirement, large surface per unit volume, low operation cost and flexibility [3,4].

Many factors affect MBR performance: hydraulic residence time (HRT), sludge retention time (SRT), temperature, feed-to-microorganism ratio (F/M), mixed liquor suspended solid (MLSS), aeration (as oxygen source and membrane fouling reducer) and biomass properties [1]. The effect of these parameters on MBR performance

and membrane fouling has been the subject of some studies [5,6]. Among these factors, HRT is one of the most influential factors since it is directly related to reactor volume and this in turn affects the capital and operational costs [7].

Membrane fouling is an important challenge in MBR applications and hence much research has been carried out to alleviate this problem [1,8]. Previous studies have shown that various parameters such as operating parameters (SRT [9], HRT [7], DO concentration) [10] and biomass properties (MLSS [11], biomass viscosity [12], extracellular polymeric substance (EPS) [13], soluble microbial product (SMP) [14], pore size [15] and foam production) [1] affect membrane fouling in aerobic MBRs. Additionally, Defrance et al. studied the contribution of various constituents of activated sludge such as suspended solids, colloids, and dissolved molecules to the resistance to filtration caused by fouling which were found to be 65%, 30% and 5%, respectively [16]. Lee et al. reported that fouling increases by increasing SRT from 20 to 60 days [17]. The effect of sludge was investigated by Meng and the results showed that EPS have a profound effect on membrane fouling [8]. Although it was reported that at high sludge concentrations more severe fouling occurs on submerged filtration membranes [18], less fouling was observed under certain conditions [19]. The relationship between membrane pore size and fouling in PVDF microfiltration membranes was investigated by Chae et al. [20]. The effect of coagulants on hollow fiber membrane fouling was also investigated, and it was shown that coagulants are more effective on fouling reduction from

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hollow fiber membranes with 0.05 μm pore size compared with those with 0.10 μm [21]. However, all of these studies investigated some effective parameters on membrane fouling with various wastewaters under different operation conditions.

As manageable operation parameters, HRT and temperature are two major factors that contribute to different treatment performances and biomass characteristics, which certainly affect membrane fouling and effluent quality in an SMBR.

To reduce membrane fouling, HRT was found to be a key parameter [22]. Huang observed that when HRT decreases, membrane fouling happens faster at constant membrane surface area [23]. Additionally, temperature can significantly affect SMBR performance. Thermophilic treatment is attractive for industries producing hot wastewaters. Several studies have been conducted on thermophilic MBRs [24–26], and MBR has been found as the most reliable system at higher temperature. However, there has to be a trade-off between the cost and the quantity of treated wastewater when appropriate HRT and temperature are being selected. From the above, it can be concluded that the effects of HRT and temperature on MBR performance still need more investigation.

The purpose of this study is therefore to examine the effects of HRT (12, 10 and 8 h) and temperature (25, 30 and 35 °C) on membrane fouling and MBR performance in a laboratory-scale SMBR using a phenolic synthetic wastewater as influent.

MATERIALS AND METHODS

1. SMBR Set Up

The SMBR in this study, as shown in Fig. 1, consisted of a glass reactor with a working volume of 5 L and an immersed hollow fiber membrane module with characteristics as given in Table 1. Synthetic wastewater kept in a steel tank was fed to the reactor by a peristaltic pump at desired flow rates. Effluent was then removed through the membrane and collected in a permeate tank via a vacuum pump. Vacuum pressure was measured by a pressure sensor and controlled accordingly. The reactor was aerated using an air pump and several porous diffusers. Steel-made junctions were used where necessary to prevent oxidation.

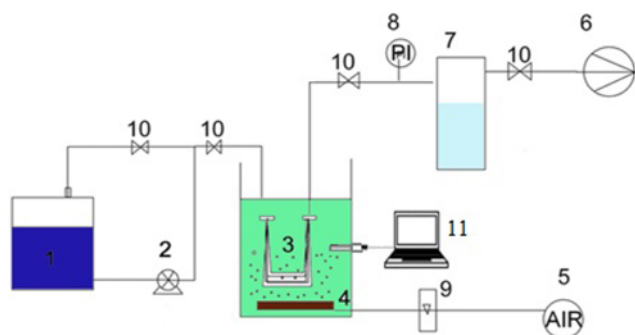


Fig. 1. Schematic of the SMBR.

- | | |
|--------------------|--------------------|
| 1. Feed tank | 7. Permeate tank |
| 2. Feed pump | 8. Pressure sensor |
| 3. Membrane module | 9. Air flow meter |
| 4. Air diffuser | 10. Valve |
| 5. Air pump | 11. DO probe |
| 6. Vacuum pump | |

Table 1. Characteristics of the hollow fiber membrane module

| Membrane type | Module type | Material | Pore size (μm) | Surface area (m^2) | Length (m) |
|------------------|-------------|----------|-----------------------------|-------------------------------|------------|
| MF, Hollow fiber | U | PVDF | 0.2 | 5×10^{-2} | 0.4 |

Table 2. Specifications of the synthetic wastewater

| Components | Concentration (mg/L) |
|-------------------------------------------|----------------------|
| Phenol | 1000 |
| KH_2PO_4 | 280 |
| K_2HPO_4 | 360 |
| NH_4Cl | 200 |
| $\text{CaCl}_2 \cdot 6\text{H}_2\text{O}$ | 67 |
| $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ | 248 |
| $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ | 0.5 |

2. Wastewater Composition

Due to the toxicity of phenolic compounds present in wastewaters of many industries, a synthetic phenolic wastewater was prepared and used as influent to the SMBR. To support microbial activities, mineral salt medium was used to prepare the synthetic wastewater [27]. To this medium phenol was added at 1,000 mg/L, which is equivalent to an influent COD of 2,380 mg/L. Specifications of the synthetic wastewater are given in Table 2. The pH and carbon-to-nitrogen (C/N) ratio of influent were adjusted at 6.9 and 20, respectively [28].

3. Inoculation and Acclimation of Activated Sludge

Activated sludge used in this study was firstly taken from Tehran Oil Refinery with an initial MLSS of about 2,000 mg/L. This sludge was previously enriched and acclimatized in presence of phenol and glucose in another SMBR using a ceramic membrane within two months to reach an MLSS of 5,000 mg/L, which is suitable for use in SMBR [29]. Thereafter, in all experiments, the SMBR with the hollow fiber membrane was employed with the appropriate initial MLSS concentration of 5,000 mg/L.

4. Experimental Procedure

To run the SMBR in continuous mode, the synthetic wastewater was fed to the reactor with a desired flow rate and the effluent was removed with the same flow rate and no sludge was removed during each experiment with infinite SRT. The observed increase in MLSS concentration from 5,000 to 11,000 mg/L was the result of both HRT and temperature variations. To monitor pH and DO, a pH meter (Lab-215, Palintest Inc.) and a DO probe (HACH, Germany) were installed in the SMBR. The values of pH and DO were approximately 6.5–7 and 2–4.5 mg/L, respectively. Firstly, a set of experiments was carried out to investigate the effect of HRT (at 8, 10 and 12 h) on the MBR performance at constant temperature of 25 °C by adjusting the feed flow rate (from 10 to 7 mL/min). The lowest HRT of 8 h was then used to examine the effect of temperature (at 25, 30 and 35 °C). Temperature was controlled using a controller/heater at about ± 1 °C. All the experiments were continuously run until reaching severe membrane fouling at transmembrane pressure of 0.5 bar. At this time, the membrane was chemically cleaned using NaOH (0.5%) followed by HCl (0.3%). The samples were daily collected

from the effluent and analyzed for phenol, COD and biomass concentrations.

5. Analytical Methods

COD and phenol concentrations were measured by spectrophotometry (Palintest, England) according to the standard method procedures described in [30] and [31], respectively. For measuring MLSS and MLVSS, six samples were taken each time. The samples were filtered through a 0.45 μm Millipore filter and dried in an oven at 105 °C for MLSS and 550 °C for MLVSS until obtaining constant weights. The average values were then reported.

RESULTS AND DISCUSSION

1. Effect of HRT

To design an MBR, it is most desirable to achieve higher phenol removal at lower HRT. Therefore, an optimum (minimum) HRT is needed to be experimentally found. In this section, the effect of HRT on MBR performance is discussed.

1-1. Phenol Removal

Phenol removals by SMBR at various HRTs are depicted in Fig. 2 at 25 °C showing fluctuations over experimental time. Therefore, to examine the effect of HRT on SMBR performance more accurately, the average phenol removal at each HRT was calculated and reported in Fig. 3. The high average phenol removals of greater than 99.5% for all HRTs (see Fig. 3) show the high capability of SMBR in phenol removal. The MF membrane completely retains the bio-

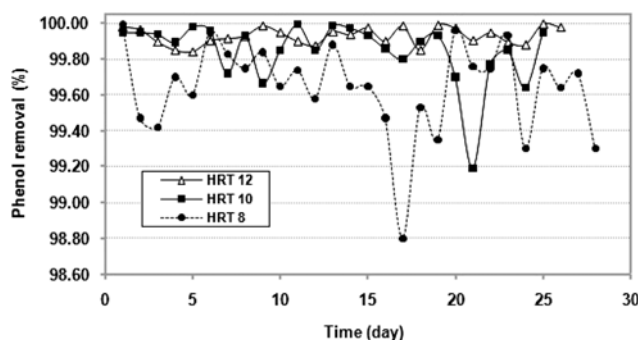


Fig. 2. Profile of phenol removal at different HRTs and constant temperature of 25 °C.

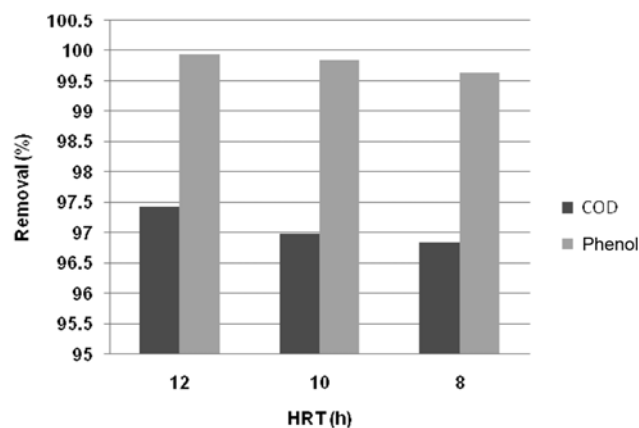


Fig. 3. Average phenol and COD removals at various HRTs at 25 °C.

mass in the mixed liquor such that effluent contains no detectable solids [33]. Therefore, MBRs can operate at high SRTs and MLSS. This in turn enhances biodegradation capability of MBR in contaminant removal compared to conventional activated sludge systems having low SRTs [1,32].

The slight increase observed in phenol removal with HRT is due to the fact that at lower HRT, wastewater has less contact time with microorganisms in SMBR which results in less phenol consumption and hence higher phenol concentration in effluent [34]. Additionally, Fig. 3 shows the average COD removals of greater than 95% in SMBR for all HRTs, and this exhibits the high performance of this system in COD removal compared to common biological COD removals of about 85-90% in wastewater treatment systems [32]. It can also be seen that phenol and COD removals are directly correlated since phenol serves as the only carbon source.

Increases in COD removal with HRT are similar to those obtained by Ren [35], Kargi [36] and Song [37].

1-2. Membrane Operation

The SMBR was first run at HRT equal to 12 h followed by 10 and 8 h. To avoid disturbing HRT results, no backwashing was applied during the experiments. However, when the membrane was severely fouled, TMP about 0.5 bar (based on membrane charac-

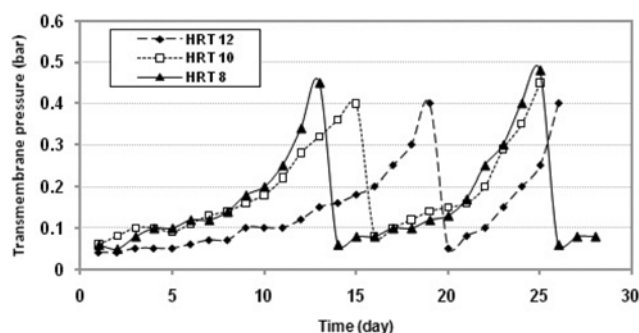


Fig. 4. Profile of TMP at different HRTs and constant temperature of 25 °C.

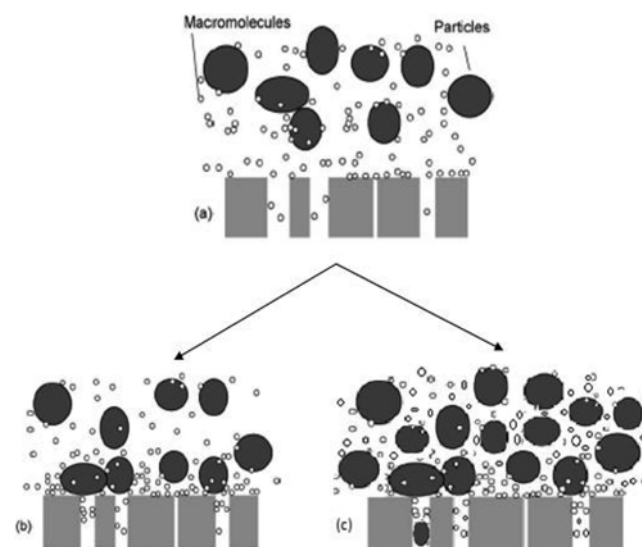


Fig. 5. Particles approaching membrane surface at (a) initial condition [35], (b) high HRT and (c) low HRT.

teristics given by the manufacturer), chemical cleaning was performed. TMP profiles at various HRTs are presented in Fig. 4.

The lowest HRT of 8 h resulted in a TMP of about 0.4–0.5 bar within 12 days while similar TMPs were reached at about 15 and 19 days for HRTs of 10 and 12 h, respectively. This shows that membrane fouling is most significant at the lowest HRT (highest effluent flow rate at same membrane surface area). This can be explained by the high velocity of blocking materials towards the membrane surface at low HRTs [33]. These materials are then absorbed by the

membrane and hence fouling occurs earlier. Figs. 5(a)–(c) illustrate the blocking materials approaching the membrane surface at initial condition, high and low HRTs, respectively.

Another aspect of effect of HRT on membrane fouling will be explained in Section 3.4.1.

2. Effect of Temperature

Due to the importance of temperature on SMBR performance, the effect of temperature was also examined in this study. Since in all examined HRTs, phenol removal was above 99.5%, which is quite satisfactory in wastewater treatment applications, the lowest HRT (8 h) corresponding to the highest effluent flow rate of 12 L/m²·h was selected for further investigations.

2-1. Phenol Removal

The profiles of effluent phenol concentration and phenol removal at 25, 30 and 35 °C are shown in Fig. 6. Results show that phenol removals were greater than 98.8, 99.5 and 99.7% at 25, 30 and 35 °C within 28, 12 and 7 days, respectively, when severe fouling was observed. Thereafter, the experiments at 30 and 35 °C were terminated.

The average phenol and COD removals are presented in Fig. 7. The average phenol removal is practically the same for all temperatures studied but shows a slight increase with temperature due to the higher microbial activity. Nonetheless, it is greater than 99.5% at all temperatures showing suitability of the MBR for toxic material removal. The average COD removal also slightly rises with temperature, which is in accordance with the results obtained by Zhang and co-workers [9]. During the experiments, COD removal was higher than 96.5%, which is quite satisfactory for wastewater treatment processes.

2-2. Membrane Operation

TMP profiles at different temperatures are depicted in Fig. 8. The membrane hydraulic flux was constant at 12 L/m²·h corresponding to HRT=8 h. Comparison of the results show that membrane was fouled earlier at higher temperature. Severe fouling (TMP about 0.5 bar) occurs within 12, 5 and 3 days at 25, 30 and 35 °C, respectively.

This study proves that temperature is also a significant operating parameter as it could change the biomass properties, compared to HRT which is an important controlling parameter particularly [9].

The effect of temperature on EPS and SMP has been studied by some researchers [9,13]. The main function of EPS is to aggregate

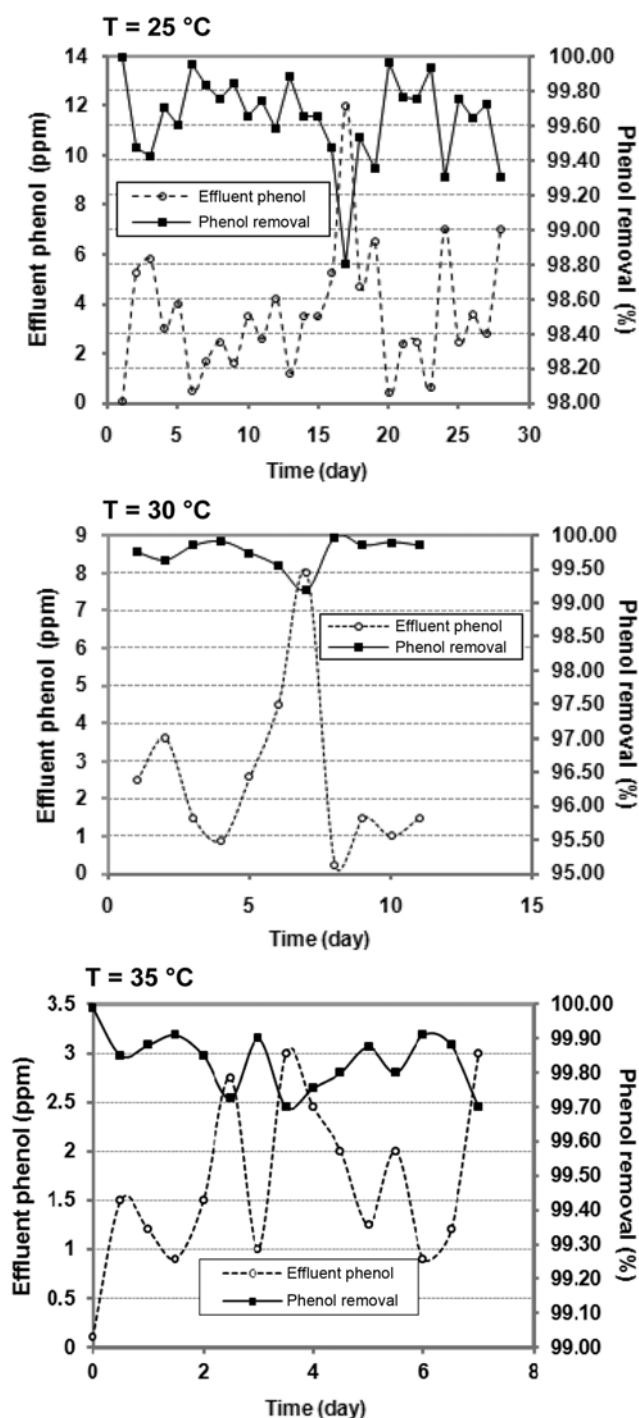


Fig. 6. Phenol concentration and phenol removal Profiles at various temperatures (25, 30 and 35 °C) and HRT=8 h.

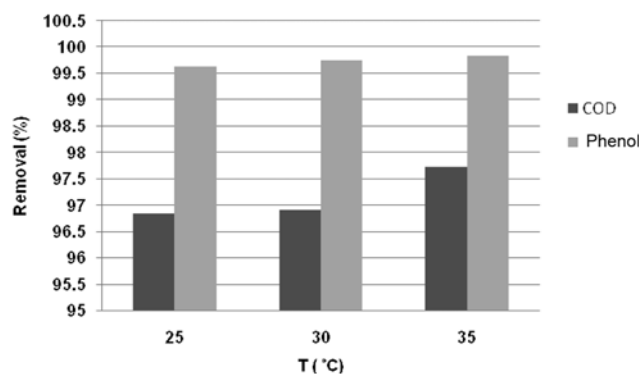


Fig. 7. Average phenol and COD removals at different temperatures and HRT=8 h.

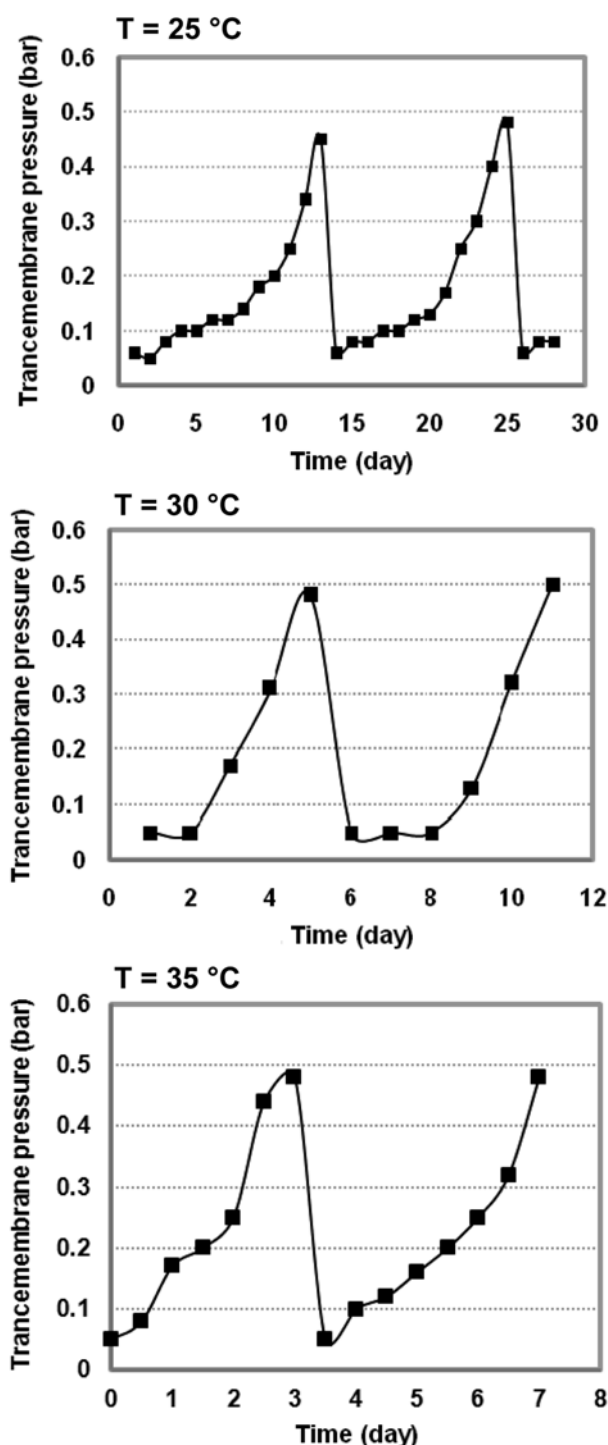


Fig. 8. TMP profiles at various temperatures (25, 30 and 35°C) and HRT=8 h (hydraulic flux of 12 L/m²·h).

the bacterial cells into flocs and biofilm and to provide a protective barrier around bacteria [13]. EPS plays a key role in membrane fouling of the submerged MBRs [38]. Vogelaar observed that EPS production decreases with temperature [39]. This can lead to lower growth rate and possible destruction of bacterial cells, resulting in SMP release into mixed liquor as shown in Fig. 9 [13]. Fouling occurs via two phenomena: internal (irreversible) and external (reversible) fouling. In internal fouling, SMP component embeds the membrane

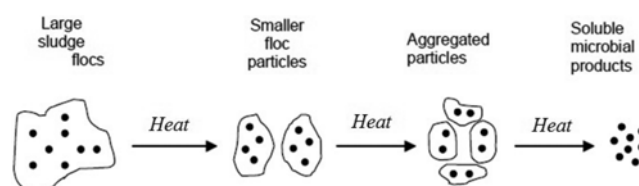
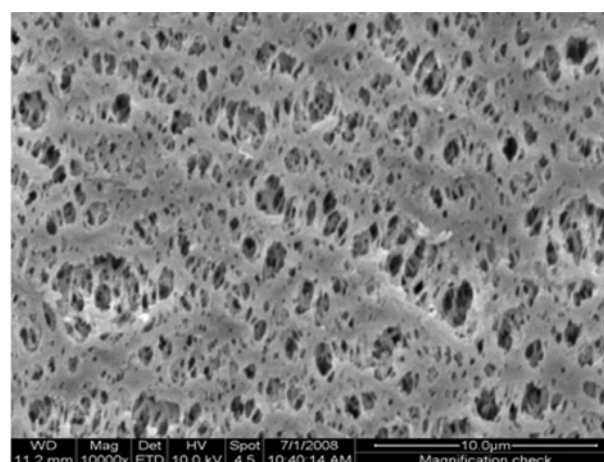
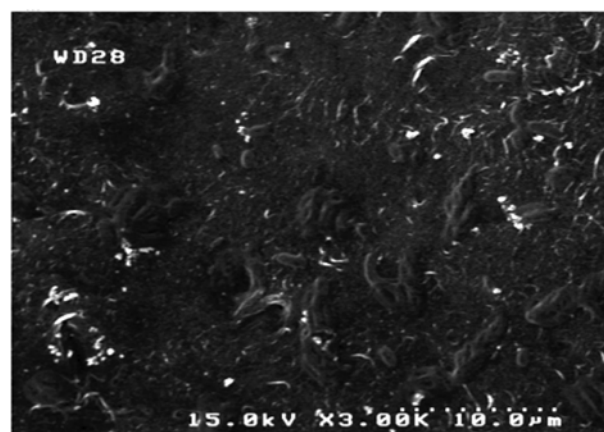


Fig. 9. Influence of heat on particle size.



(a)



(b)

Fig. 10. FE-SEM images of hollow fiber membrane surface, (a) cleaned and (b) fouled.

pores, and in external fouling, SMP components form a gel matrix which attaches the membrane surface. On the other hand, increasing temperature results in the membrane bores widening [9]. By increasing temperature, these phenomena act together to develop the membrane fouling.

3. Field Emission Scanning Electron Microscopy (FE-SEM) Measurement

Observation of the membrane with FE-SEM (Hitachi S-4160, accelerating voltage of 15 kV) was also carried out. FE-SEM was operated under dry mode (10^{-7} Torr, dry membrane). FE-SEM images of cleaned and fouled membrane surfaces are illustrated in Fig. 10. Membrane pores of the clean membrane can be clearly observed in Fig. 10(a), while fouled membrane is covered with a slim gel layer as observed in Fig. 10(b). Once the gel layer is developed, it

is difficult to remove the layer from the membrane surface by routine aeration and this increases the TMP. At this point, the membrane needs to be cleaned chemically ex-situ.

FE-SEM cross-sectional images of cleaned and fouled membrane are presented in Fig. 11. Before cleaning, the pores alongside the external surface of the membrane are more fouled, while after cleaning pore blockage becomes clearer.

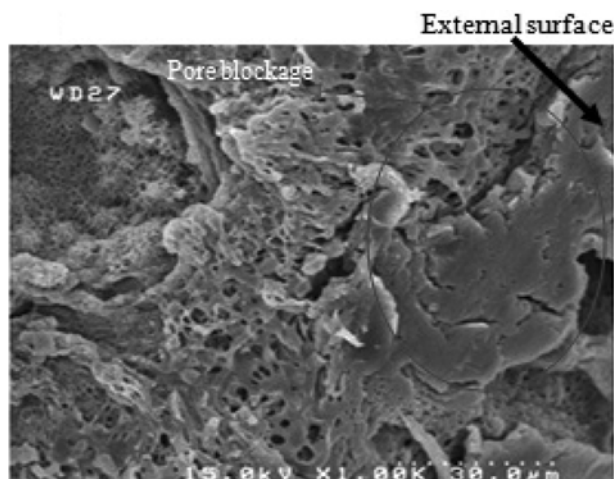
4. Effect of HRT & Temperature on Biomass Growth

The number of microorganisms in a biological environment is one of the key factors in organic material removal. Although increasing the number of microorganisms can enhance the removal efficiency, DO is reduced as a result of increased oxygen uptake rate, and this affects the cell life cycle and terminates the operation [40].

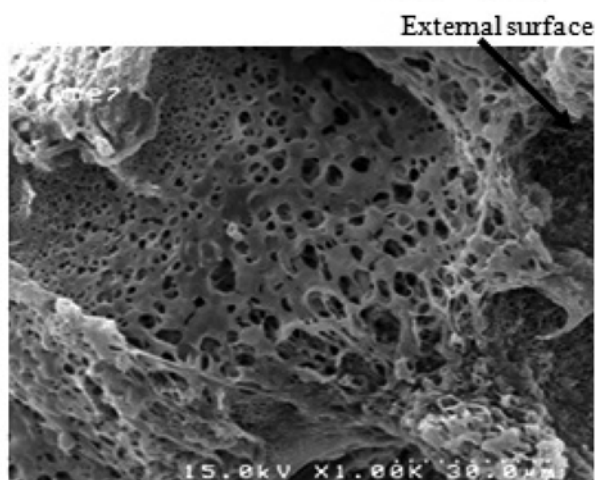
In all experiments, the SMBR was employed with the appropriate initial MLSS concentration of 5,000 mg/L.

4-1. Effect of HRT on MLSS & MLVSS

Variation of MLSS and MLVSS with time is presented in Fig. 12, at $T=25^{\circ}\text{C}$ for different HRTs. Similar trend is observed at all HRTs with an initial increasing stage followed by a second stage of fluctuation.



(a)



(b)

Fig. 11. FE-SEM images of hollow fiber membrane cross-section, (a) cleaned and (b) fouled.

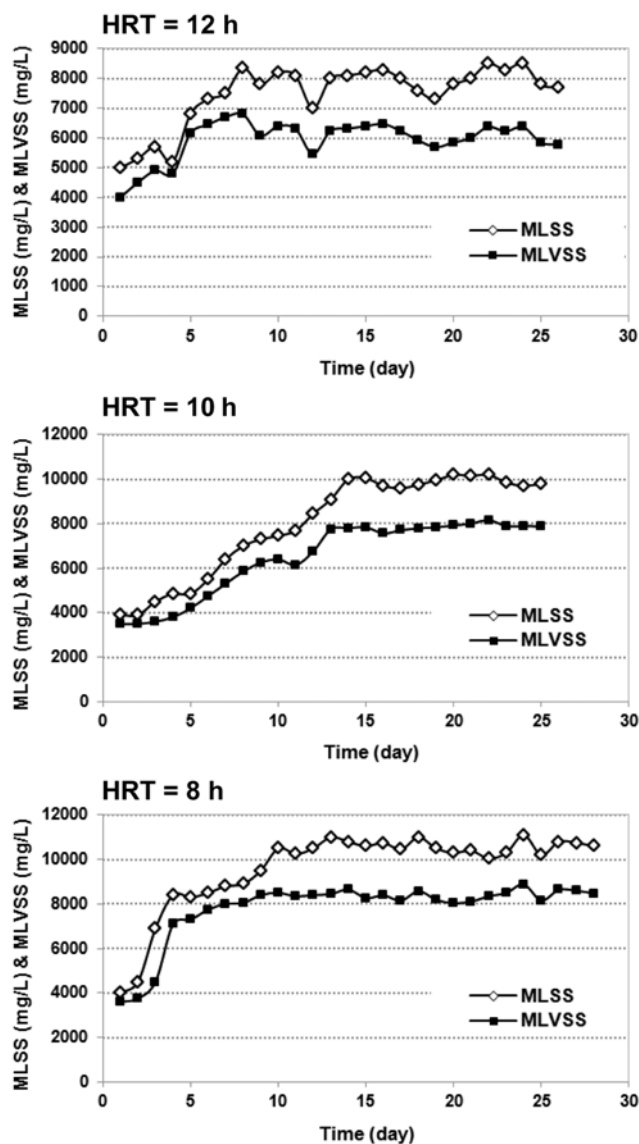


Fig. 12. MLSS and MLVSS profiles at various HRT (12, 10 and 8 h) and $T=25^{\circ}\text{C}$.

Table 3. Variation of MLVSS with HRTs at $T=25^{\circ}\text{C}$

| HRT (h) | MLVSS (mg/L) |
|---------|--------------|
| 12 | 6200 |
| 10 | 7900 |
| 8 | 8500 |

Variation of MLVSS concentration with HRT is given in Table 3. As can be seen, the highest MLVSS value is achieved at the lowest HRT. This phenomenon occurs due to the higher loaded nutrients into bioreactor at lower HRTs, which can support microbial growth.

Sludge concentration reduces with HRT. This means that at Low HRT of 8 h membrane is exposed to a more concentrated culture, which can further contribute to membrane fouling.

4-2. Effect of Temperature on MLSS & MLVSS

Variation of MLSS and MLVSS with time at different temperatures for HRT=8 h is shown in Fig. 13.

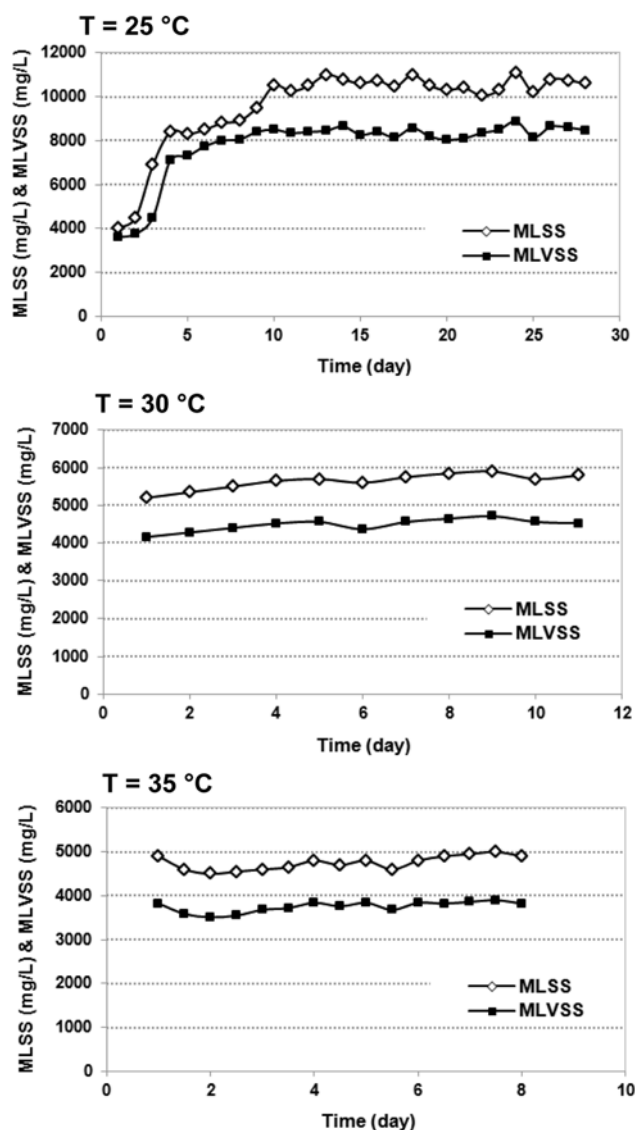


Fig. 13. MLSS and MLVSS profiles at various temperatures (25, 30 and 35 °C) and HRT=8 h.

Table 4. Variation of MLVSS with temperature at HRT=8 h

| T (°C) | MLVSS (mg/L) |
|--------|--------------|
| 25 | 8500 |
| 30 | 4600 |
| 35 | 3900 |

Table 4 illustrates variation of MLVSS concentration with temperature. As observed, by increasing temperature, biomass concentration

(MLVSS) significantly decreases.

This can be explained by the fact that at high temperatures, microorganisms utilize a large amount of energy to maintain their vital function, which can increase biomass stability. As a result, synthesis of new cellular materials ceases and this results in reduction of biomass concentration. These results are in agreement with those obtained in previous studies [13,41].

Another point to consider is the high phenol removal efficiency at higher temperature of 35 °C alongside lower concentration of MLVSS. Achieving efficient performance using SMBRs usually needs high level of sludge concentration [1], which can cause operational difficulties. Therefore, by increasing temperature while using SMBRs, one can benefit from the reduced production of sludge.

5. Variation of pH and DO with Time

Variation of pH and DO are important operational factors in SMBRs. Table 5 shows the values of pH and DO for all combinations of HRTs and temperatures used in this work.

During the experiments, it was observed that pH varies in the range of 6 to 7.5, which is similar to that reported in other studies [40,42]. With respect to DO, despite the fact that air flow rate was almost constant at all temperatures, it was observed that DO concentration decreases with temperature. This can be explained by the fact that increasing temperature results in lower oxygen solubility in the mixed liquor. In addition, microorganism activity increases at higher temperature, which results in higher oxygen uptake by microorganisms [1,38].

CONCLUSIONS

A submerged aerobic MBR was firstly operated at three HRTs (12, 10, 8 h) and HRT (8 h) was then used to investigate the effect of temperature (at 25, 30, 35 °C). SMBR performance with respect to removal efficiency, fouling, MLSS, MLVSS, DO, pH, was then examined leading to the following conclusions:

- At all HRTs, phenol and COD removals are above 99% and 95%, respectively, which shows the high performance of SMBR in removing toxic organic compounds.
- Although fouling happens earlier at increased effluent flow rate (decreased HRT), time differences are not very significant. Therefore, HRT=8 h equivalent to the highest effluent flow rate (at fixed bioreactor volume) which is desirable from operational aspects was selected.
- Increasing temperature causes more fouling; however, phenol removal remains above 99.5%.
- With increasing temperature, the amounts of MLSS and MLVSS show considerable reduction. Hence, the system can reach high removal with less sludge output, which is desirable for biological systems.

This study showed that SMBR can be used to efficiently treat

Table 5. DO and pH variations at different HRTs and temperatures

| Variables | Conditions | HRT (h) at T=25 °C | | | T (°C) at HRT=8 h | | |
|-----------|------------|--------------------|---------|-------|-------------------|---------|---------|
| | | 12 | 10 | 8 | 25 | 30 | 35 |
| DO (mg/L) | | 3-4.5 | 2.5-4.8 | 2.9-4 | 2.9-4 | 2.5-3.5 | 2.2-3 |
| pH | | 6-7.5 | 6.2-7 | 6-7.5 | 6-7.5 | 6-7 | 6.5-7.5 |

phenolic wastewater at a range of flow rates and temperatures, among which HRT=8 h and T=25 °C are the preferred operating conditions resulting in high flow rate and moderate membrane fouling.

NOMENCLATURE

EPS : extracellular polymeric substance
 HRT : hydraulic retention time [h]
 MBR : membrane bioreactor
 MF : microfiltration
 MLSS : mixed liquor suspended solid [mg/L]
 MLVSS : mixed liquor volatile suspended solid [mg/L]
 SMBR : submerged membrane bioreactor
 SMP : soluble microbial product
 SRT : sludge retention time [day]
 TMP : transmembrane pressure [bar]
 UF : ultrafiltration

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