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# Membrane fouling in a submerged membrane bioreactor treating high strength municipal wastewater

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**ABSTRACT**

Studying the influence of the operating parameters on membrane fouling is important in fouling control. This paper presents the interaction effects of the sludge retention time (SRT), organic loading rate (OLR) and feed temperature ($T_f$) on membrane fouling. A submerged membrane bioreactor (SMBR) was operated under a constant flux (11.1 l/m$^2$ h), with different SRT, $T_f$ and OLR. A synthetic wastewater representative of high strength municipal wastewater was fed to the bioreactor. Three different levels (low, medium and high) of SRT, $T_f$ and OLR were studied. These were 25, 30 and 35 days for SRT, 20, 30 and 40°C for $T_f$ and OLR of 1.73, 4.03 6.82 kg COD/m$^3$.d. The sustainable time ($t_{sus}$) was defined as the time at which the rate of suction pressure started to increase rapidly. $t_{sus}$ was found to increase with low SRT and high $T_f$. A higher OLR resulted in higher mixed liquor suspended solids (MLSS), however it did not cause a faster membrane fouling. Applying higher aeration rate enabled a longer sustainable time to be obtained. Sustainable time $t_{sus}$ was found to be well correlated with MLVSS/MLSS with an $r^2$ of 0.995. The range of MLVSS/MLSS tested varied from 74.3 to 82.3% at which $t_{sus}$ decreased 2.9-fold (from 175 to 60.5 h).

*Keywords*: Membrane bioreactor (MBR); Membrane fouling; Sustainable time ($t_{sus}$); Sludge retention time (SRT); Feed temperature ($T_f$); Organic loading rate (OLR)

## 1. Introduction

Membrane bioreactor (MBR) combines the biological degradation process by activated sludge with a direct solid–liquid separation by membrane filtration. MBR systems can be classified into two major groups according to their configurations. The first group is known as an integrated MBR which exposes the outer skin of the membranes to the content of the bioreactor, i.e., the membrane is internal (known as submerged MBR) [1–3]. The second configuration is the recirculated (external) MBR. In this system, the mixed liquor is recirculated through a membrane module that is outside the bioreactor [4]. Submerged MBRs have been used worldwide with the emergence of less expensive and more resilient polymeric

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membranes along with lower pressure requirements and higher permeate fluxes [5]

The MBR has many advantages over conventional wastewater treatment processes. These include the excellent effluent quality due to retention of all suspended matter and most soluble compounds within the bioreactor [6], small footprint and reactor requirements; the possibility of retaining all bacteria and viruses results in sterile effluent and consequently eliminating extensive disinfection [7]; the potential of operating the MBR at very high sludge ages without having the obstacle of settling and allows high biomass concentrations in the bioreactor. Consequently, higher strength wastewater can be treated and lower biomass yields are realized [8].

Membrane fouling can be simply defined as the decrease of flux over time. This phenomenon is commonly considered as a weakness point in MBR applications. Membrane clogging in the MBR process might be (a) external fouling which results from biofilm growth, or adsorption or deposition of foulants on the top surface of the membrane and (b) internal fouling that takes place at the pore entrances or within the internal pore structure of the membrane. Adsorption is used here to mean an interaction between foulants and membranes [9].

Due to the complex heterogeneity of the filtered matrix, investigation of foulant species and fouling mechanisms in MBRs is more complicated than other MF or UF processes, as reviewed in depth by Le-Clech et al. [10]. Membrane characteristics (materials, pore size, hydrophobicity/hydrophilicity and roughness), fluid characteristics (feed composition, floc properties and biomass activity) and operation conditions (both biological and hydraulic) can all contribute to determine the best possible configuration which can be significantly different even for very similar systems [11].

In this study, a synthetic wastewater representative of high strength municipal wastewater was prepared and acclimatized with the sludge supplied from Tesco Damansara domestic wastewater treatment plant, Malaysia, for 20 days after which the COD removal efficiency was stable at about 80%. This paper investigates the interaction effects of the operation parameters namely, SRT, $T_f$ and OLR on the membrane fouling in MBR treating high strength municipal wastewater. The results of ten experiments carried out using laboratory scale MBR are presented.

## 2. Materials and methods

### 2.1. Experimental setup

A schematic diagram of the experimental unit is shown in Fig. 1. The bioreactor consists of two compartments; aerated and non-aerated with a working volume of 20.4 l (15.3 l for aerated compartment and 5.1 l for non-aerated compartment). A microfiltration membrane module was immersed in the aerated compartment for filtration. An air pump was used to aerate the reactor through a diffuser fixed at the bottom of the reactor. Table 1 shows the specifications of the microfiltration membrane used in the study.

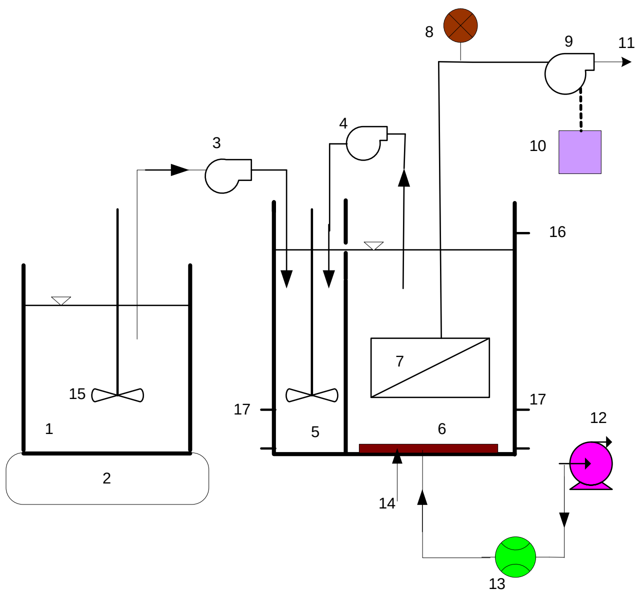


Fig. 1. Layout of the experimental setup. 1 Feeding tank, 2 Water bath/low temperature bath circulator, 3 Feeding pump, 4 Circulating pump, 5 Non-aerated compartment, 6 Aerated compartment, 7 Membrane module, 8 Pressure gauge, 9 Suction pump, 10 Programming time controller, 11 Final effluent, 12 Oxygen supply aerator, 13 Air flow meter, 14 Air diffuser, 15 Mixer, 16 Level controller, 17 Sampling port.

Table 1
Specifications of the membrane module

| | |
|---|---|
| Membrane material | Polyethylene |
| Outer diameter, μm | 540 |
| Inner diameter, μm | 350 |
| Pore size, μm | 0.4 |
| Surface area, $m^2$ | 0.2 |
| Manufacturer | Mitsubishi Rayon (Japan) |

### 2.2. Feed wastewater

Synthetic wastewater was used in this study instead of actual wastewater, to control the variation of nutrient concentrations as otherwise found in raw wastewater. The chemical concentrations used to prepare the synthetic wastewater are as in Table 2 [12]. However, in this study, the stock solution prepared was five times more concentrated than that of Jin et al. [12]. The stock solution was kept in the refrigerator at 4°C, ready-made for daily use. The stock solution was diluted with distilled water to a desired COD concentration. Laguna clay suspension was added at a rate of 165 ml per 1 l of the

Table 2
Composition of synthetic wastewater (mg/l)

| Composition | Concentration (mg/l) Jin et al. [10] | Concentration used in the study (mg/l) |
|---|---|---|
| Glucose | 670–1080 | 5400 |
| Glutamic acid | 285–460 | 2300 |
| $CH_3COONH_4$ | 220–350 | 660 |
| $NaHCO_3$ | 750–3000 | 5000 |
| $NH_4Cl$ | 33–53 | 165 |
| $KH_2 PO_4$ | 60 | 300 |
| $K_2HPO_4$ | 80 | 400 |
| $MgSO_4 .7H_2O$ | 33 | 165 |
| $FeCl_3.6H_2O$ | 2 | 10 |
| $CaCl_2.2HO$ | 20 | 100 |
| NaCl | 25 | 125 |

synthetic wastewater to increase the suspended solids. Laguna clay suspension was prepared as described in Mohammed et al. [13].

### 2.3. System operation

The system was operated at different SRT, $T_f$ and OLR for three different levels of SRT, $T_f$ and OLR. These levels were 25, 30 and 35 days for SRT; 20, 30 and 40°C for $T_f$ and OLR of 1.73, 4.03 6.82 kg COD/m$^3$.d. Water bath and low temperature bath circulator were used to control the feed temperature to the required level. Initial hydraulic retention time (HRT) was fixed at 8 h. The feed water was pumped to the MBR through a peristaltic pump. Wastewater was circulated from the aeration compartment to the anoxic compartment through the circulated pump at the same rate of the inflow to achieve the denitrification process. The permeate was obtained through suction with the peristaltic pump (Fig. 1) in an intermittent mode of operation. A filtration period of 8 min was followed by a filtration pause of 2 min. The most stable membrane performance could be achieved through this mode of operation [14]. A portion of sludge was removed from the reactor daily according to the operating SRT. The mixed liquor suspended solids (MLSS) and dissolved oxygen (DO) concentration were measured regularly and when the DO dropped lower than 1 mg/l, the aeration rate was increased. Flux and suction pressure were measured hourly during each run.

## 3. Results and discussion

### 3.1. Development of flux and suction pressure under different operating conditions

In this study, the experimental trials were categorized according to the wastewater strength, low, medium and high as shown in Table 3. The MBR was operated under a constant flux (11.1 l/m$^2$.h) throughout the experiments.

The development of permeate flux ($J_p$) and suction pressure ($SP$) with time under the different operating conditions carried out, is presented in Figs. 2–4. It is observed that all figures show similar trends for both $J_p$ and $SP$. The figures show a constant $J_p$ and a slight increase in $SP$ and that the rate of $SP$ increases with time. This phenomenon is caused by membrane fouling. It is the result of accumulation of rejected particles on the top of the membrane — external fouling, or deposition and adsorption of small particles or macromolecules at the pores or within the internal pore structure of the membrane — internal fouling [15].

In recent years scientists have interpreted the fouling mechanisms in MBR in many ways. An initial stage of fouling during constant flux operation was found to be caused by the strong interactions between the membrane surface and the EPS present in the mixed liquor [16].

Passive adsorption of colloids and organics has been observed even for zero-flux operation and before initiation of any deposition mechanism [17]. Another study based on passive adsorption exposed that the hydraulic resistance due to this process was almost independent of tangential shear. The initial adsorption was found to contribute some 20–2000% of the clean membrane resistance, mainly depending on pore size [18]. However, its

Table 3
Category of the experiments based on synthetic wastewater strengths

| Parameter | Low strength (Trial no. 1, 4, 6, 9) | | | Medium strength (Trial no. 2 and 10) | | | High strength (Trial no. 3, 5, 7, 8) | | |
|---|---|---|---|---|---|---|---|---|---|
| | Min. | Max. | AV. | Min. | Max. | AV. | Min. | Max. | AV. |
| COD, mg/l | 520 | 760 | 619 | 1320 | 1680 | 1500 | 2280 | 2760 | 2437 |
| BOD, mg/l | 360 | 604 | 473 | 832 | 1117 | 965 | 1538 | 2140 | 1871 |
| $NH_3$-N, mg/l | 10.9 | 32.6 | 19.4 | 33.0 | 45.0 | 41.5 | 46.6 | 83.3 | 55.4 |
| TSS, mg/l | 53 | 190 | 130 | 170 | 236 | 198 | 175 | 380 | 295 |
| VLR, kg COD/m$^3$ d | 1.2 | 1.95 | 1.73 | 3.06 | 4.50 | 4.03 | 5.2 | 7.4 | 6.82 |

AV: Average

contribution to the overall resistance was found to become negligible once filtration was conducted [15]. Study done on a test cell equipped with direct observation through membrane technology, applied cross-flow but with zero-flux. Flocs were visually observed to temporarily land on the membrane [17]. This was described as a random interaction process rather than proper cake formation phenomenon. During the movement of some flocs across the membrane, biological aggregates detached and left residual marks of smaller flocs or EPS material. This would make attachment of the biomass approaching the membrane surface easier, colonising the separation surface and contributing to the next.

*Stage 1— slow fouling*: After the initial stage, most of the membrane surface is expected to be covered by soluble microbial products (SMP), leading to higher attachment propensity of biomass particles and colloids. More adsorption and deposition of organics may also occur during this stage. The adsorption may occur not only at the pores but also on the whole surface. Therefore, biological flocs may initiate cake formation without affecting the permeability in this stage. Over time, this phenomenon would worsen [16]. Consequently; slight increase in *SP* was observed in this stage.

*Stage 2 — SP jump*: As some areas or pores of the membrane are fouled, the flux in those locations would decrease, leading to redistribution of the overall permeate productivity to the less fouled membrane areas or pores. The increase of local flux for these areas would exceed the critical flux (sustainable flux). If the filtration is maintained, severe fouling is generally obtained. Consequently a significant increase in TMP or jump would occur [17].

### 3.2. Sustainable time

In order to define a standard parameter for the comparative evaluation of the trials, $t_{sus}$ was estimated as the time at which the rate of suction pressure started to increase rapidly (Figs. 2–4).

Guglielmi et al. [11] estimated the time of sustainability ($t_{sust}$) as the last point at which a correlation factor for an exponential fitting of the TMP curve was more than 95%. In the present study, the correlation factor ($r^2$) for exponential fitting of the SP curve ranged between 0.922 and 0.959 as shown in Figs. 2–4. This shows that the current $t_{sus}$ estimation and that of Guglielmi et al. [11] are comparable to some extent.

The sustainable time for each trial was determined from its corresponding flux–*SP* curve and the results of the ten trials are shown in Fig. 5, from which the different sustainable times are observed.

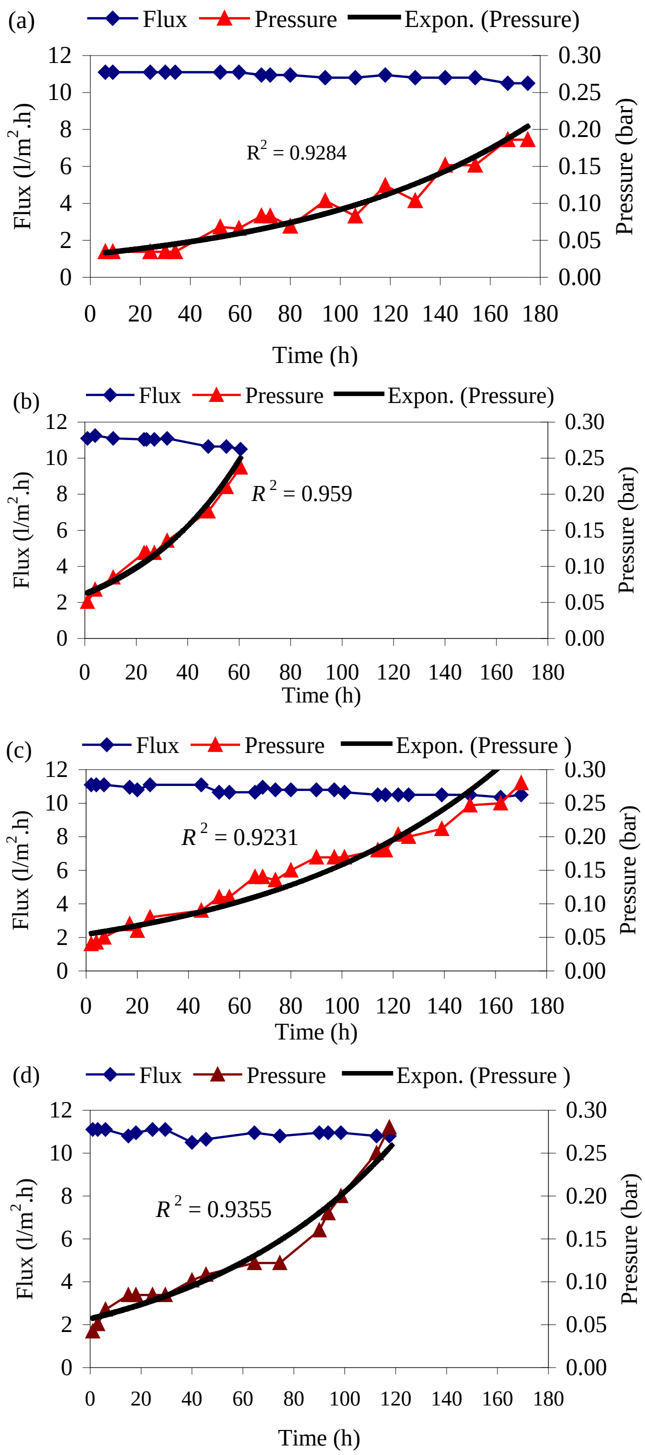


Fig. 2. Membrane flux and suction pressure vs. time for low strength wastewater: (a) Trial 1, (b) Trial 4, (c) Trial 6 and (d) Trial 9.

### 3.3. Interaction effects of solid retention time and feed temperature on the flux sustainability

Trials 1, 4, 6 and 9 present the results of sustainable time for the low wastewater strength trials (Table 3). Fig. 5 shows that the sustainable time of trials 1, 4, 6 and 9 is 175, 60.5, 167 and 117.5 h, respectively. The operating conditions of these trials are shown in Table 4.

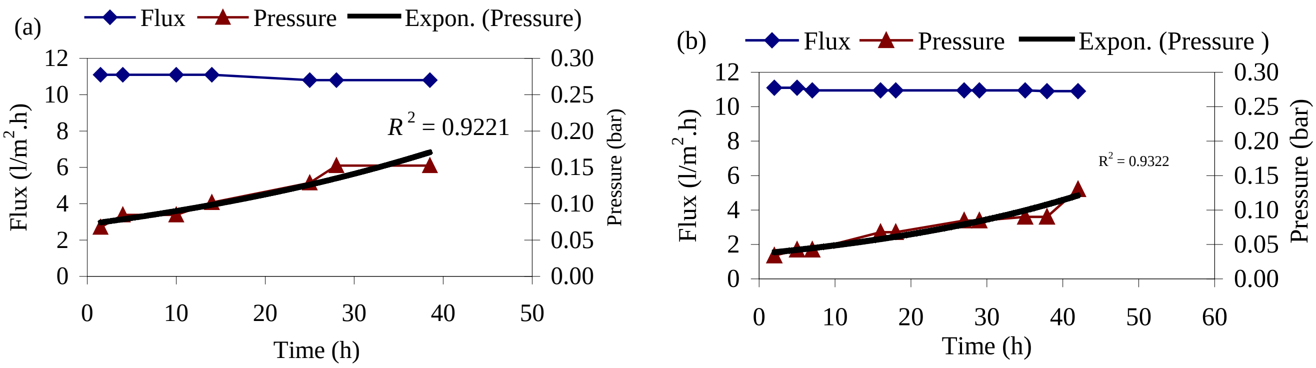


Fig. 3. Membrane flux and suction pressure vs. time for medium strength wastewater: (a) Trial 2 and (b) Trial 10.

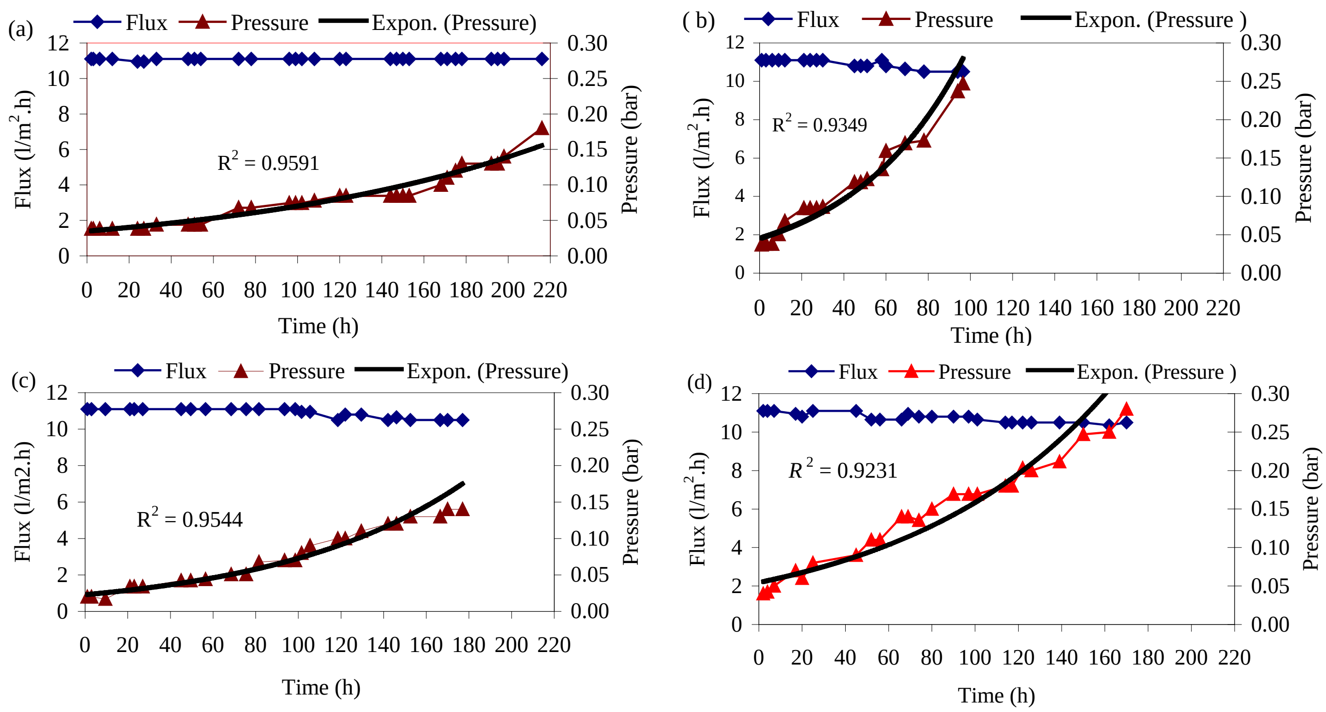


Fig.4: Membrane flux and suction pressure vs time for high strength wastewater; (a) Trial 3, (b) Trial 5, (c) Trial 7 and (d) Trial 8.

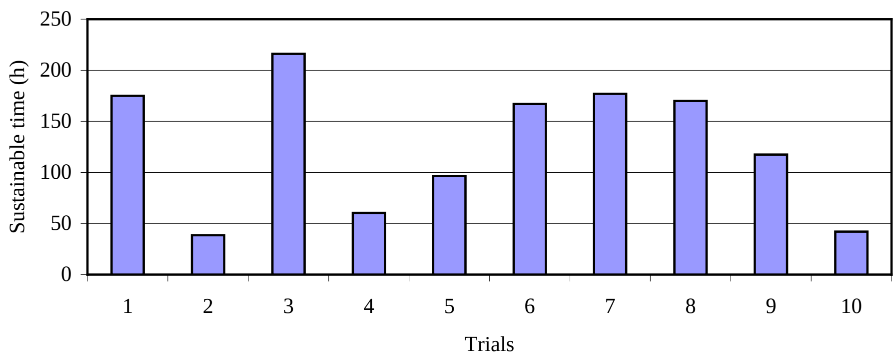


Fig. 5. Sustainable time for the different trials.

Table 4
Experimental design

| Full factorial design | | | |
|---|---|---|---|
| Factors: 3 | | Base design: 3, 8 | |
| Blocks: none | | Center pts (total): 2 | |
| All terms are free from aliasing | | | |
| Data matrix (randomized) | | | |
| Run | A | B | C |
| 1 | – | + | – |
| 2 | o | o | o |
| 3 | + | + | – |
| 4 | – | – | + |
| 5 | + | – | + |
| 6 | – | – | – |
| 7 | + | – | – |
| 8 | + | + | + |
| 9 | – | + | + |
| 10 | o | o | o |
| | OLR | $T_f$ | SRT |

Note: + represents the higher value
– represents the lower value
o represents the medium value

The low SRT, which results in low MLSS concentration coupled the high $T_{f,}$ which leads to low viscosity are favorable for longer sustainable time (175 h). In trial 6, although the low SRT extended the sustainability, but low temperature reduced the duration, therefore resulting in less $t_{sus}$ (167 h). The sustainable time in trial 9 was reduced by high SRT. However, it was increased by the high $T_f$ and the resultant $t_{sus}$ obtained was 117.5 h. The lowest $t_{sus}$ (60.5 h) was obtained in trial 4, which was carried out under low $T_f$ and high SRT, both of which had negative effects on MBR filtration.

These results can be explained by the fact that within the same organic loading rate, lower SRT meaning lower biomass concentration (MLSS) can be produced in the bioreactor. This parameter has shown a complex interaction with MBR fouling, but controversial findings of the effects of this parameter have also been reported. If the other biomass characteristics are neglected, some authors reported that increasing MLSS concentration had resulted in a negative impact (higher TMP or lower flux) on the MBR hydraulic performance [18,19]. However, others reported positive impacts [20] or insignificant impacts [21]. Rosenberger et al. [22] found that while an increase in MLSS had decreased the fouling rate at low MLSS concentration (<6 g/l), more fouling was expected if MLSS concentration was above 15 g/l. SRT also affects the EPS characteristics in the MBR, which was found to be the major fouling parameter [3,23,24] A clear decrease of EPS levels was observed for the extended SRT, but this reduction became negligible for SRT greater than 30 days [1].

Secondly, flux was found to have increased with increasing temperature for the same concentration of MLSS and the reason was that the sludge viscosity decreased at a higher temperature [25]. Moreover, the back transport of deposited particles from the membrane surface to the water depends on the shear force induced by air scouring and liquid turbulence, both of which will be accelerated in the liquid of a lower viscosity [26].

The sustainable time for the high strength wastewater is indicated by trials 3, 5, 7 and 8. Their $t_{sus}$ recorded ranged between 96.5 and 216 h; trial 3 (216 h), trial 7 (177 h), trial 8 (170 h) and trial 5 (96.5 h) as shown in Fig. 5. It is observed that the longest sustainable time was obtained at high $T_f$ and low SRT, followed by $t_{sus}$ at low $T_f$ and low SRT, $t_{sus}$ at high $T_f$ and high SRT and $t_{sus}$ at low $T_f$ and high SRT (Table 4). The trend of this result is similar to that of the low strength wastewater trials. This confirms the rightness of interpretation for the interaction effect of SRT and $T_f$ that introduced in the above paragraph.

In general, within the range of the operating parameters adopted in this study, it can be concluded that lower SRT would result in a longer sustainable time, while higher feed temperature would also lead to the same result.

### *3.4. The effect of organic loading rate (OLR) on flux sustainability*

Organic loading rate has a very significant effect on the MLSS concentration, the concentration of MLSS in this study was found to increase slowly for the low strength, moderately for the medium strength and rapidly for the high strength wastewaters.

Table 5 shows the sustainable time for the two trials (low and high OLR) conducted under the same conditions (same SRT and $T_f$). It indicates that the sustainable time of the high OLR was longer than that of the low OLR. Higher MLSS and higher mixed liquor viscosity, as discussed in section 3.3, should increase the membrane fouling rate thereby shortening sustainable time. However the results obtained showed longer sustainable time for high OLR. The reason is the higher aeration rate generated when MBR operated at high OLR. For the high OLR (high MLSS concentration), a higher aeration rate was needed to achieve the DO level required for the microorganism's activities. Therefore, the aeration rate range for both high and low OLR trials were 20–60 l/min and 20-40 mg/l respectively (Table 5).

Bubbles near the membrane surface will induce local shear transients and liquid flow fluctuations. The tangential shear at the membrane surface prevents large particles deposition on the membrane surface [16]. Another effect of aeration on MBR performance is that it causes fibre lateral movement in hollow fiber configurations [27]. A

Table 5
Comparison of the sustainable time between the low and high OLR trials

| Trials | | Aeration rate (l/min) | | Sustainable time (h) | |
|---|---|---|---|---|---|
| Low OLR | High OLR | Low OLR | High OLR | Low OLR | High OLR |
| Trial 1 | Trial 3 | 20-25 | 30-60 | 175 | 216 |
| Trial 4 | Trial 5 | 30-40 | 40-50 | 60.5 | 96.5 |
| Trial 6 | Trial 7 | 20-25 | 20-60 | 167 | 177 |
| Trial 9 | Trial 8 | 20-40 | 30-55 | 117.5 | 170 |

recent study conducted by Ji and Zhou [28] showed that the aeration rate had direct control over the quantity and composition of polymeric compounds (EPS) in the biological flocs and finally the ratio of protein/carbohydrate deposited on the membrane surface. Their conclusion was that the cycle length increased with increasing aeration rates, and this finding is supportive of the present study.

The findings discussed in this section conclude that higher organic loading rate had resulted in a higher MLSS concentration, however, it did not cause faster membrane fouling. Applying higher aeration rate enabled a longer sustainable time to be obtained.

### *3.5. Correlation between flux sustainability and MLVSS/MLSS ratio*

Fig. 6 shows the correlation between sustainable time and the average MLVSS/MLSS ratio for the low organic loading trials. It is observed that the sustainable time correlated ($r^2$ was 0.995) with the MLVSS/MLSS ratio. For the 8% range of MLVSS/MLSS ratio tested (from 74.3 to 82.3%), the sustainable time decreased approximately 2.9-fold (from 175 to 60.5 h). This can be associated with the higher MLVSS/MLSS ratio which implies higher percentage of organic matter (including biomass cells, EPS and organic colloids) produced in the mixed liquor.

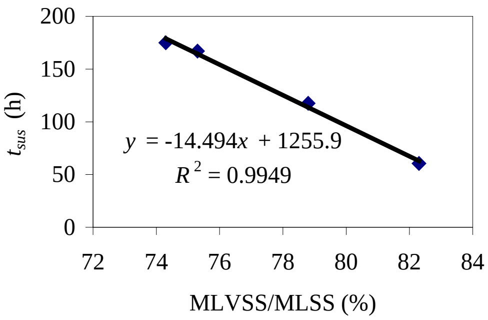


Fig. 6. Sustainable time vs. MLVSS/MLSS ratio for low strength wastewater.

## 4. Conclusions

The interaction effects of SRT, $T_f$ and OLR on membrane fouling were investigated with the sustainable time ($t_{sus}$), as the time at which the rate of pressure started to increase rapidly, was used as a measure to compare the results of the different trials. In general, within the same range of OLR, $t_{sus}$ increased with decreasing SRT and increasing $T_f$. It was found that higher OLR resulted in higher MLSS concentration however, did not cause a faster membrane fouling. Applying higher aeration rate enabled a longer sustainable time to be obtained.

It was found that $t_{sus}$ decreased as MLVSS/MLSS increased with a correlation of $r^2$ = 0.995. Thus, within the 8% range of MLVSS/MLSS ratio tested (from 74.3 to 82.3%), the sustainable time decreased approximately by 2.9-fold from 175 to 60.5 h.

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