The effect of mixed liquor suspended solids (MLSS) on biofouling in a hybrid membrane bioreactor for the treatment of high concentration organic wastewater

A. Damayanti, Z. Ujang, M. R. Salim and G. Olsson

ABSTRACT

Biofouling is a crucial factor in membrane bioreactor (MBR) applications, particularly for high organic loading operations. This paper reports a study on biofouling in an MBR to establish a relationship between critical flux, \( J_c \), mixed liquor suspended solids (MLSS) (ranging from 5 to 20 g L\(^{-1}\)) and volumetric loading rate (6.3 kg COD m\(^{-3}\) h\(^{-1}\)) of palm oil mill effluent (POME). A lab-scale 100 L hybrid MBR consisting of anaerobic, anoxic, and aerobic reactors was used with flat sheet microfiltration (MF) submerged in the aerobic compartment. The food-to-microorganism (F/M) ratio was maintained at 0.18 kg COD kg\(^{-1}\) MLSS d\(^{-1}\). The biofouling tendency of the membrane was obtained based on the flux against the transmembrane pressure (TMP) behaviour. The critical flux is sensitive to the MLSS. At the MLSS 20 g L\(^{-1}\) the critical flux is about four times lower than that for the MLSS concentration of 5 g L\(^{-1}\). The results showed high removal efficiency of denitrification and nitrification up to 97% at the MLSS concentration 20 g L\(^{-1}\). The results show that the operation has to compromise between a high and a low MLSS concentration. The former will favour a higher removal rate, while the latter will favour a higher critical flux.

Key words | biofouling, flux, membrane bioreactor, MLSS, TMP

INTRODUCTION

Palm oil mill has significantly contributed to the generation of wastewater in Malaysia. As the world’s largest producer and exporter of palm oil, Malaysia has more than three million hectares of oil palm cultivated area. More than 500 palm oil mills have been built and operated for the past 40 years (Damayanti et al. 2010). Palm oil processing is carried out in mills where oil is extracted from the oil palm fruit bunch (Vijayaraghavan et al. 2007). Extraction of crude palm oil needs large quantities of water, contributing to about 50% of the POME. POME is characterized as a thick brownish liquid containing high amounts of total solids (\( \approx 40 \) g L\(^{-1}\)), oil and grease (\( \approx 8 \) g L\(^{-1}\)), COD (\( \approx 45 \) g L\(^{-1}\)), BOD (\( \approx 25 \) g L\(^{-1}\)), ammonia nitrogen (\( \approx 0.1 \) g L\(^{-1}\)), and pH of 3.5\( \pm \)0.1 (Ahmad et al. 2008).

In recent years, membrane technology has been used basically for low strength wastewater like domestic wastewater in aerobic conditions similar to the conventional activated sludge process (Hamdzah et al. 2005; Ujang et al. 2007). The membrane bioreactor (MBR) has been successfully used for treatment of municipal and industrial wastewaters (Alaboud 2009). The research reported in this paper has been conducted under different operating conditions using a hybrid MBR (HMBR) with a combination of anaerobic, anoxic, and aerobic reactors to treat POME. A sustainable process performance in the last aerobic tank was obtained at different levels of MLSS concentration 5 to 20 g L\(^{-1}\). The membrane replaces the function of the settling tanks that is conveying the supernatant to the water body as well as...
returning the solids back to the reactor (Qin et al. 2007). Internal biofouling due to the adsorption of dissolved matter into the membrane pores and pore blocking is considered irreversible and is generally only removed by chemical cleaning. There are principally three strategies for limiting biofouling in submerged MBR process systems: maintaining the permeate flux below the critical flux, $J_c$, increasing the membrane aeration, and employing physical or chemical cleaning (Razak et al. 2007). Under constant-TMP or constant-flux conditions, the fouling mechanism will increase hydraulic resistance and the trans membrane pressure will be increased as permeate flux will be declined, as further studied by Qin et al. (2007).

Many studies about biofouling at MBR in different MLSS, for several example Chang & Kim (2005) using synthetic wastewater, showed which cake resistance ($R_c$) decreased as MLSS concentration (between 0.09–3.7 g L$^{-1}$) decreasing. Another study compared attached growth (MLSS between 0.1–2 g L$^{-1}$) and suspended growth (MLSS 3 g L$^{-1}$) microorganisms in MBR using municipal wastewater showed the rate of attached growth system 7 times higher affected the biofouling rate compare with suspended growth (Lee et al. 2001). Therefore this study was carried out to investigate performance and biofouling tendency at four different MLSS concentrations between 5 to 20 g L$^{-1}$ in a hybrid MBR treating high strength wastewater, like POME. Thus the study has two aims: (1) to observe the biofouling through the critical flux, $J_c$, which was determined using the flux step method and (2) to investigate the performance of the organic and nitrogen removal in the system at different MLSS levels.

**MATERIALS AND METHODS**

**POME samples and pretreatment process**

The characteristics of raw POME collected from Felda Bukit Besar Palm Mill, Johor, Malaysia are shown in Table 1. Upon sampling the temperature of POME was measured at approximately 80°C±5°C. POME samples used in this study were stored at 4°C. The operating temperature was 25°C (room temperature in a tropical country). The POME was mixed in the reactor using a stirrer according to Hofvendahl et al. (1999) for anaerobic compartment and Ramalho et al. (2004) for anoxic compartment, for agitation speed 120 rpm or Newton number 4 to maximize the process. Finally the POME was collected in the feed tank to study the performance of different levels of MLSS. The F/M was calculated using Equation (1) based on measurements taken from the experiment. The values of F/M range from 0.7 to 1 kg BOD$_5$ kg MLSS$^{-1}$ day$^{-1}$ (Metcalf and Eddy 2004).

\[
\frac{F}{M} = \frac{Q S_0}{V X} \tag{1}
\]

where $F/M =$ food to microorganism ratio

$S_0 =$ influent concentration (ML$^{-3}$)

$Q =$ flowrate (L$^3$T$^{-1}$)

$V =$ volume (L$^3$)

$X =$ biomass concentration (ML$^{-3}$).

**Laboratory scale trials**

The schematic process flow diagram for the lab scale HMBR is shown in Figure 1. A specification of the HMBR system is given in Table 2. For acclimatization the HMBR system was seeded with biological sludge taken from the wastewater treatment plant of the Felda Bukit Besar Palm Mill, Johor Bahru. The experiments were carried out at room temperature 25°C. The system was operated continuously (24-h) during this study, with acclimatization taking placed within two months and a total study duration of six months. Trials for different MLSS levels were conducted to obtain biofouling conditions. The HRT of the anaerobic, anoxic, and aerobic reactors, as shown in Table 2, were 11, 7, and 8 hours respectively. The flat sheet membrane (15cm × 10cm) used

<table>
<thead>
<tr>
<th>Symbol</th>
<th>POME Values$^a$</th>
<th>Sewage$^{b,c}$</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>5.6</td>
<td>7</td>
</tr>
<tr>
<td>Tot COD</td>
<td>45</td>
<td>0.60</td>
</tr>
<tr>
<td>TS (Total Solid)</td>
<td>43</td>
<td>0.80</td>
</tr>
<tr>
<td>TDS (Total Dissolved Solid)</td>
<td>35</td>
<td>0.23</td>
</tr>
<tr>
<td>SS (Suspended Solid)</td>
<td>8.2</td>
<td>0.56</td>
</tr>
<tr>
<td>NVSS (Non Volatile Suspended Solid)</td>
<td>4.0</td>
<td>0.25</td>
</tr>
<tr>
<td>NO$_3$</td>
<td>0.10</td>
<td>0.01</td>
</tr>
<tr>
<td>NO$_2$</td>
<td>0.30</td>
<td>0.06</td>
</tr>
<tr>
<td>NH$_4^+$</td>
<td>0.20</td>
<td>0.03</td>
</tr>
<tr>
<td>Total Nitrogen</td>
<td>0.50</td>
<td>0.01</td>
</tr>
</tbody>
</table>

*Note:
$^a$This study,
$^b$Nor-Anuar et al. (2007),
$^c$Ujang et al. (2008)
All parameter’s units in g L$^{-1}$ except pH.
in this study had been kindly donated by Kubota, Inc. and was vertically submerged in the bioreactor. The size of the membrane bioreactor is 24cm × 10cm × 90cm. The height between the membrane module and the base of the reactor is 30 cm. The TMP was measured using an online system by installing a pressure transducer. For the aeration, the compressed air supplied individually and the flowrate has been used is 12 LMH. Recirculation flow rate in this study was 1:3.

The system is operated for acclimatization to the new media was about one a half months before starting experiments. The sludge divided into 4, for MLSS study 5, 10, 15 and 20 g L\(^{-1}\). After that, the sludge placed in cool room. Then the system operated using lowest MLSS first, 5 g L\(^{-1}\), acclimatized again around one to two weeks until steady state phase, and finished it in one month. After that, the sludge was replaced from reactor, using new sludge (from cool room) which have acclimatized before, but it is still need acclimatized again around one to two weeks until steady state phase, using MLSS 10 g L\(^{-1}\), until finished in one month. The same step as MLSS 15 and 20 g L\(^{-1}\). We excess sludge withdrawal and stationary MLSS for same SRT, 70 days. All step study finished in four months excluded acclimatization in two months. Flux 11 Lm\(^{-2}\) h\(^{-1}\) applied for operating condition in HMBR.

During the studies, the fouled membranes were removed from the aeration reactor and cleaned with a sponge followed by water flush cleaning. The membranes were then soaked in a 0.5% ppm NaOCl and left overnight (Ujang et al. 2005). After this, the cleaned membranes were ready to be used again in the process (Ujang et al. 2005).

**Critical flux evaluation**

The flux step method is used to determine the critical flux (Le-Clech et al. 2003). The critical flux measured at the start for each period. This carried out by increasing values of permeate flux and recording the relative TMP variations. For each flux step two TMP values were considered: the initial TMP, corresponding to the initial sudden increase of the filtration resistance, and the final TMP, i.e. the TMP at the end of the step (Le-Clech et al. 2003). From these two TMP values two parameters connected to fouling can be evaluated, the average TMP and the rate of change of the TMP (\(\frac{d\text{TMP}}{dt}\)). Any flux with \(\frac{d\text{TMP}}{dt} \geq 0.5\) is defined as the critical flux (Bottino et al. 2009). By using resistance series model as in Equation (2), \(R_{\text{tot}}\) or total filtration resistance then could be calculated (Chang & Kim 2005):

\[
R_{\text{tot}} = R_m + R_c + R_f
\]

where \(R_{\text{tot}}\) is the total filtration resistance, \(R_m\) is the intrinsic membrane resistance (obtained from pure water permeability measurements), \(R_c\) is the cake resistance, and \(R_f\) is the fouling resistance due to irreversible adsorption and pore plugging (Bottino et al. 2009). \(R_m\) and \(R_{\text{tot}}\) were calculated according to Darcy’s law below:

\[
R = \text{TMP} \mu^{-1} \text{f}^{-1}
\]
where $\mu$ is the fluid viscosity and $J$ is the flux. Permeate flux ($J$) applied from 0 to 40 Lm$^{-2}$h$^{-1}$ for critical flux test, 15 minute for one value constant. The critical flux measured at the start each test period of MLSS. Each MLSS concentration used new membrane.

**Samples analysis**

Laboratory experiments were carried out in the Environmental Engineering Laboratory, UTM. All experimental analysis was conducted according to Standard Methods (APHA 1998) within three hours after sampling. MLSS concentrations were regularly measured. Chemical Oxygen Demand, NO$_3$-N, NO$_2$-N, NH$_4$-N, and PO$_4$-P were analysed using a spectrophotometer (HACH/DR 4000). The measurements were carried out in duplicate according to HACH testing procedures. The average values were taken for different MLSS concentrations between 5 and 20 g L$^{-1}$. Dissolved oxygen concentration, temperature and pH were monitored using an on-line measurement system, and the biotransformation was observed using on-line respirometry. The DO probe used in this work was the WTW Oxil oxygen meter. It was calibrated with pure water before being used in the membrane bioreactor.

**RESULTS AND DISCUSSION**

**Behaviour of membrane critical flux in MBR operation**

Figures 2 and 3, and Table 3 show the influence of different MLSS concentrations on the TMP and biofouling behaviour.

The values in Table 3 are calculated using Equation (3). The results clearly demonstrate that the membrane biofouling rate increases with an increase in the MLSS concentration. The results obtained indicate that the MLSS concentration has a significant influence on the trans membrane pressure (TMP).

**Figure 2** shows the flux vs. TMP behaviour, during the ascending and descending run of membrane with POME at different MLSS concentrations. The differences between two runs were significant.

During the ascending run, the flux first proportionally increased with an increase in the TMP and leveled off, more quickly at higher MLSS concentrations, due to membrane biofouling. The detrimental effect of fouling on the membrane flux is also apparent by observing the behaviour of the membrane flux as a function of the applied TMP during the descending run.

**Figure 3** shows the trend of $d\text{TMP}/dt$ as a function of the flux at MLSS concentrations 5–20 g L$^{-1}$ with Y-error bar 5%.

![Figure 2](image1.png)  
**Figure 2** | TMP and $J$ behavior during ascending and descending runs at different MLSS concentrations with Y error bar 5%.

![Figure 3](image2.png)  
**Figure 3** | Trend of $d\text{TMP}/dt$ (mbar/sec) as a function of the flux at MLSS concentrations 5–20 g L$^{-1}$ with Y-error bar 5%.

<table>
<thead>
<tr>
<th>No</th>
<th>MLSS (g/L)</th>
<th>$J_c$ (Lm$^{-2}$h$^{-1}$)</th>
<th>$R_{tot}$ ($10^{12}$ m$^{-1}$)</th>
<th>$R_m$ ($10^{12}$ m$^{-1}$)</th>
<th>$R_c + R_f$ ($10^{12}$ m$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>5</td>
<td>9.2</td>
<td>1.73</td>
<td>0.19</td>
<td>1.54</td>
</tr>
<tr>
<td>2</td>
<td>10</td>
<td>7.1</td>
<td>2.04</td>
<td>0.19</td>
<td>1.85</td>
</tr>
<tr>
<td>3</td>
<td>15</td>
<td>4.7</td>
<td>2.83</td>
<td>0.19</td>
<td>2.64</td>
</tr>
<tr>
<td>4</td>
<td>20</td>
<td>2.5</td>
<td>3.90</td>
<td>0.19</td>
<td>3.71</td>
</tr>
</tbody>
</table>
cake formation and fouling occurred during the treatment process. Table 3 demonstrates that higher MLSS will lead to higher $R_{tot}$. The $R_m$ value led to a significant increase of the total resistance, since biofouling and cake formation take place, especially at high MLSS concentration (Bottino et al. 2009). Unlike Bottino et al. (2009), Chang & Kim (2005) showed that the $R_m$ value not always led to a significant increase of the total resistance, especially at low MLSS concentration. This is in line also with Chang & Kim (2005) that found that $R_c$, cake resistance, will decrease as MLSS between 3.7 to 0.09 g L$^{-1}$ decrease.

Chang & Kim (2005) also showed that higher MLSS concentrations lead to higher higher $R_{tot}$. The rate of TMP increase was found to generally increase with increasing MLSS concentration at each flux tested because of higher suspended solids in the reactor (Bottino et al. 2009). Furthermore, at each MLSS concentration, $dTMP/dt$ increased with increasing flux-step. In addition, Rosenberger & Kraume (2002) who found that higher SS concentration affected lower F/M ratio and also lower EPS concentration, led to better filterability.

**COD, NO$_3$-, NO$_2$-, and NH$_4^+$ removal efficiency at different flux and MLSS**

COD, NO$_3$-, NO$_2$-, and NH$_4^+$ removal efficiencies at different MLSS are shown in Figure 4. Towards the end of the experimental period the feed COD concentration had an average of 40.5 g L$^{-1}$, while the effluent concentration was kept in the range 200 to 350 mg L$^{-1}$ depending on the POME source quality and MLSS used in the MBR. These mean values here is continuous with acclimatized sludge from lab scale hybrid MBR. This effluent quality need to be treated further to meet effluent quality standard (Environmental Quality Regulation 2009).

Figure 4 also demonstrates a COD removal efficiency of up to 99%. The NO$_3$-N effluent concentration depends on the POME source quality and the average removal rate is up to 97%. This is comparable with other studies, where NO$_3$ removal using aerobic treatment was up to 95% (Kumbasar 2009) and 95% NH$_4^+$ removal (Sarioğlu et al. 2009). In Figure 4 it is shown that the rate of removal efficiency generally increases with increasing MLSS concentration at each flux tested because of higher biomass activity working in the reactor (Alaboud 2009). Furthermore, at each MLSS concentration, $dTMP/dt$ increased with increasing flux-step. This is why balancing the biofouling tendency and effluent quality is needed to reduce operation and maintenance costs in a wastewater treatment plant (Ujang et al. 2008). It is not a big difference but it is important for the effluent quality at high MLSS concentrations. However it is also important to consider the biofouling effect at high MLSS concentrations.

**CONCLUSIONS**

This study has indicated that the membrane biofouling rate increases with an increasing MLSS concentration, as observed through the critical flux analysis and extensive experimental results for POME. During the experiments of increasing MLSS the flux first increased proportionally with the increase in TMP and levelled off, more quickly at higher MLSS concentration, due to membrane biofouling. The detrimental effect of fouling on the membrane flux is also apparent by observing the behaviour of the membrane flux as a function of the applied TMP during the descending run. The rate of change in the TMP was found to generally increase with increasing MLSS concentration at each flux tested. The study also showed that the removal increased with increasing MLSS concentrations. In general, the results from this paper indicate the need to balance the biofouling tendency and the effluent quality to reduce operation and maintenance costs in wastewater treatment plants, for high strength wastewater such as POME.

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