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Transient Performance of MBR with Flux Enhancing Polymer Addition

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Transcript of the document:

The polymeric membrane performance enhancer (MPE50) was applied in two parallel submerged membrane bioreactors to test its effect on membrane fouling. Although the flux enhancer showed significant benefit, the effect of dosage termination caused a dramatic drop in performance relative to the control. The MBRs were compared in three carefully controlled runs performed over 155 days. The total organic carbon (TOC), EPS, and particle size data were collected at regular intervals during the entire operation. In run 1, after a start-up period of 62 days, a stable state was achieved in both MBRs. In run 2, a one-off 400 mg/L MPE and 15 mg/L MPE daily maintenance dosage were applied in one reactor for 60 days. The “critical flux” determined by the flux stepping test was increased by 50% from 20 LMH to 30 LMH after the addition of MPE. Long-term experiments were implemented at three separate levels of constant flux, namely 20, 30, and 40 LMH. At the flux of 20 LMH (below the “critical flux” of both reactors with and without MPE), the performance in both reactors was similar. At the flux of 30 LMH (above the “critical flux” of the MBR without MPE but below the “critical flux” of the MBR with MPE), the addition of MPE was found to mitigate membrane fouling. At a flux of 40 LMH (above the critical flux for both reactors), the TMP rise in the MBR with MPE was slower than the MBR without MPE, although the performance of both MBRs was poor. The performance differences correlated with the increase in the particle size distribution of the activated sludge as well as the low level of SMP and EPS (mainly polysaccharides) concentrations in the supernatant during steady state. In run 3, after terminating the MPE addition, the membrane performance was found to be significantly worse than the MBR without MPE, which suggests that the continuous maintenance dose of MPE is essential upon MPE application to the MBR.

Keyword  flocculant; membrane bioreactor (MBR); membrane fouling; performance enhancer (MPE50); polymeric membrane

INTRODUCTION

Submerged membrane bioreactors (sMBRs) have been widely applied in wastewater treatment and water reclamation technology (1). The main limitation of the technology is membrane fouling which results in frequent membrane chemical cleaning and a shorter membrane lifespan. Operating at low fluxes is a popular strategy to mitigate fouling, but this will also incur higher capital cost because of the low efficiency of membrane usage. A major focus of recent research has been to develop new technology to reduce fouling and increase operating flux.

In the MBR, the activated sludge is mixed with the feed and in contact with the membrane. Generally, it comprises biological particles such as bio-flocs formed by flocculated growing bacteria, dispersed bacterial cells, protozoa, rotifers, and soluble molecules, such as organic and inorganic compounds either introduced from the raw wastewater or produced during biomass growth and decay. The state of the activated sludge will strongly affect the membrane performance under specific hydrodynamic conditions.

Sludge particles of different sizes foul the membrane via different mechanisms (2). The direct observation through the membrane (DOTM) technique has confirmed that small particles deposit preferentially on the membrane surface in tangential flow filtration (13). “Critical flux” is mainly controlled by the small particles in the mixture. Below the critical flux, no particles deposit on the membrane and above the critical flux, particles deposit on the membrane due to the transmembrane pressure (TMP), and may become irreversible foulants (4,5). It is recognized that in MBRs the concept of “critical flux” is debatable, as some degree of fouling appears to occur at any flux (6). However, we take “critical flux” to be that of the dominant foulant (biofloc) and it can be estimated by flux-stepping and TMP transients (2,7). At sub-critical flux, fouling is mainly contributed by soluble macromolecules in the supernatant of the mixed liquor, which can be sub-divided into extracellular polymeric substances (EPS) and soluble microbial products (SMP). Their concentrations are known to have a significant effect on membrane fouling under sub-critical conditions (8,9).

In order to decrease the concentrations of EPS and SMP, as well as to increase the particle size, a number of...
Different approaches have been pursued. These include prolonging the sludge retention time to decrease the EPS and SMP concentrations, using granular activated sludge, bio-PAC to increase the particle size etc. (10–13). An alternative is the use of additives to flocculate the supernatant macromolecules and increase the particle size distribution. Previous studies have shown that the addition of flocculants to a feed stream prior to microfiltration can mitigate membrane fouling in tangential flow filtration (14–16). In a MBR study by Lee et al. (17), alum addition was applied in a lab-scale submerged membrane bioreactor and a reduction in fouling was observed. When alum was added, the particle size was increased by the coagulation of small colloids and the total organic carbon (TOC) in the reactor was maintained at a low concentration. A recent study cited the use of membrane performance enhancers (MPE) (a polyelectrolyte-based flocculant recently developed by Nalco) in full- and pilot-scale MBR plants. When the MPE was dosed at 400 mg/L, the average membrane flux was reported to increase by 50–178% and the supernatant polysaccharide level reduced in the mixed liquor in the long-term operation of a pilot-scale MBR (5,18,19).

Previous studies have demonstrated that flocculant addition can improve membrane fouling in submerged MBRs. However, the biomass properties were not monitored under long-term, stable MBR operation. Most of the previous studies reported results from batch or pilot-scale experiments, and no comparison was made using parallel reactors operating under the same conditions. In this work, two carefully controlled lab-scale MBRs under steady state conditions were used to compare the effect of operation with and without MPE on the biomass. Particle size, distribution of EPS in supernatant and biofloc, Specific Oxygen Uptake Rate (SOUR) of the two types of biomass and their membrane fouling propensity were investigated and compared. These results provide further insights into the effects of flocculation on the biomass and hence the mitigation of membrane fouling. In addition, the response of the treated MBR to the termination of the flocculent was monitored and showed unexpected results.

**MATERIAL AND METHODS**

**Experimental Setup**

The experimental MBR system (Fig. 1) comprised of 2 bioreactors (30 L aerated tank) with submerged flat sheet microfiltration (MF) membrane modules (Kubota, 0.12 m² per panel and pore size of 0.2 µm). Concentrated simulated municipal wastewater was continuously pumped into the bioreactor at a constant rate, while tap water was provided as a supplement to the bioreactor through a solenoid valve controlled by a level sensor to maintain constant level in the bioreactor. In this way, the concentrated feed (see Table 1) was diluted approximately 4.8 times by the tap water. The permeate pumps were controlled by flow meters and computer controlled systems to keep the permeate flowrate constant. Each channel between the flat sheet membrane modules had separate air diffusers at the bottom of the bioreactor. The transmembrane pressure (TMP) was recorded using a Cole-Parmer high accuracy (±0.13 kPa) pressure transducer in the suction line.

**Experimental Conditions**

The 2 membrane bioreactors (MBR 1 and MBR 2) were operated in parallel for 155 days at 30 days SRT. MBR 1 served as the control reactor with zero MPE addition throughout the entire operation. MBR 2 was seeded with MBR 1 sludge and served as the test reactor with MPE.
additive addition. Both the MBRs were fed with simulated municipal wastewater at an organic load of 0.6–0.7 kg TOC/m²·day (1.5 kg COD/m²·day). Figure 2 shows the change in MLSS during the operating period. Table 2 shows the operating conditions for each reactor.

MBR 1 and 2 had a 62 days start-up period (for 62 days – run 1) to achieve steady state at a SRT of 30 days. From the 62nd day to the 122nd day (for 60 days – run 2), MPE was added to MBR 2 to reach a concentration of 400 mg/L to modify the activated sludge. Subsequently, MBR2 was dosed with 15 mg/L MPE on a daily basis to compensate for the MPE loss during sludge wasting. From the 122nd day to the 155th day (for 33 days – run 3), the addition of the MPE in MBR 2 was ceased.

When required, the membrane modules were cleaned by washing with tap water and submerging in 5–10% NaClO solution for 8 hours.

Analytical Materials and Methods

Analytical methods from the “Standard Methods for the Examination of Water and Wastewater” were adopted for the measurement of the mixed liquor suspended solids (MLSS) in the bioreactors (20). The supernatant samples were prepared by centrifuging the mixed liquor sample from the bioreactor twice at 4000 rpm for 10 minutes. The particle size distribution of the biomass was measured by a particle sizer (MALVERN Mastersizer HYDRO2000SM). The EPS extraction method followed that reported by Zhang and Liu (21,22). Supernatant EPS was physically extracted without adding any chemical extractant, simply by centrifugating (4000 G) at 4000 rpm for 10 minutes followed by high-speed centrifugation (20000 G) for 20 minutes; the pellet of the sample was resuspended with distilled water and the pellet EPS extraction followed the “formaldehyde plus NaOH extraction methods”. The total EPS was the sum of the supernatant EPS and pellet EPS. TOC was measured by a SHIMADHU TOC-VCSH.

RESULT AND DISCUSSIONS

Start-Up Period (Run 1)

Biomass Characteristics During the Start-Up Period

To determine the effect of adding MPE on membrane fouling, two types of stable biomass were obtained in MBR 1 and 2 after 62 days of operation. MBR 1 was the original MBR that has been running for 1.5 years. The design of the aeration tank of MBR 2 and 1 were the same. MBR 2 was seeded with the biomass of MBR 1 to an initial concentration of 3.50 g/L MLSS. Thereafter, 1 L of the discharged biomass from MBR 1 was added daily for

<table>
<thead>
<tr>
<th>TABLE 1</th>
<th>Composition of the concentrated synthetic wastewater (diluted approximately 4.8 x as feed)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nutrient</td>
<td>mg/L</td>
</tr>
<tr>
<td>Glucose</td>
<td>800</td>
</tr>
<tr>
<td>Meat extract</td>
<td>150</td>
</tr>
<tr>
<td>Peptone</td>
<td>200</td>
</tr>
<tr>
<td>KH₂PO₄</td>
<td>35</td>
</tr>
<tr>
<td>MgSO₄</td>
<td>35</td>
</tr>
<tr>
<td>FeSO₄</td>
<td>20</td>
</tr>
<tr>
<td>Sodium acetate</td>
<td>600</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>TABLE 2</th>
<th>Operating conditions of MBR 1 &amp; 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>MBR 1</td>
<td>MBR 2</td>
</tr>
<tr>
<td>SRT (days)</td>
<td>30</td>
</tr>
<tr>
<td>HRT (hour)</td>
<td>6</td>
</tr>
<tr>
<td>MPE dose (ppm)</td>
<td>0</td>
</tr>
<tr>
<td>Reactor temperature (°C)</td>
<td>24–26</td>
</tr>
<tr>
<td>DO in the biomass solution (mg/L)</td>
<td>3–4</td>
</tr>
<tr>
<td>Aeration intensity (m³/m².h)</td>
<td>0.75</td>
</tr>
<tr>
<td>Permeate flux (L/m².h)</td>
<td>20</td>
</tr>
<tr>
<td>pH</td>
<td>7–8</td>
</tr>
</tbody>
</table>
two weeks. After 15 days of operation, the biomass concentration in MBR 2 stabilized at around 9.28 g/L and biomass was discharged regularly to maintain a constant SRT of 30 days. Figure 2 shows the variations of the MLSS concentrations in MBR 1 and 2 variations during the start-up period. After 62 days of operation, the parameters such as total EPS and EPS/MLSS, EPS in the supernatant, TOC of permeate, TOC of the supernatant and particle size reached a stable state. The trends of these parameters are shown in the run 1 section of Figs. 3–7 respectively. The trends showed fluctuations during the first 30 days of operation and a stable state was reached in the subsequent 32 days. The average values are summarized in Table 3.

Fouling Propensity During the Start up Period

Although the biomass in both reactors were in a similar state (in terms of the parameters described in Table 3) in run 1, the fouling intensity of MBR 1 and 2 in run 1 were investigated to confirm a fair fouling comparison in run 2. A new KUBOTA flat sheet membrane module was immersed in each reactor. The fluxes were set at 20 L/m².h for both reactors. The TMP variations are shown in Fig. 8. From the 35th to 60th day in run 1, the slope of the TMP rise (dTMP/dt) was almost identical at 0.4 kPa/day in MBR 1 and 2, indicating similar membrane fouling propensity in the two parallel MBRs.

Application of MPE to MBR 2 (Run 2)

Biomass Characteristics of the Flocculated and Control Activated Sludges in MBR 1 and 2

In run 2, the permeate TOC was stable in both reactors with similar average permeate TOC concentration of 2.6 ± 0.2 mg/L (Fig. 5), indicating similar organic removal. From Fig. 6, the supernatant TOC of the MBR without and with MPE were 10.3 ± 4.0 and 6.7 ± 2.4 mg/L.
respectively. In consideration of other variability factors, this difference may not be particularly significant. In comparison with run 1, both MBR1 and MBR2 are noted to have stabilized in run 2 resulting in lower supernatant TOCs.

In Fig. 3, the total EPS in both reactors were similar with a relative average value of approximately 0.34 g/L in run 2. However, the average supernatant EPS value showed a slight difference in this run, being 11.0 ± 5.0 and 7.8 ± 4.5 mg/L for the MBR without MPE and with MPE respectively (Fig. 4). The EPS of the flocculated activated sludge by MPE was also slightly lower than that of the non-flocculated activated sludge but the difference is assumed to be negligible. The total EPS/MLSS in both reactors was the same with a relative average value around 35.0 mg/g MLSS. In summary, the EPS analysis for run 2 showed that the total EPS and EPS/MLSS of both MBRs were very low.

Figure 7 reveals a significant difference in the particle size in MBR 1 and 2 in run 2. The particle size was increased significantly by the addition of MPE. The d (0.1) and d (0.5) are the particle sizes based on the volume percentage, below which 10% and 50% of the distribution fall. The d (0.5) can be a good indicator for the large particles distribution. The addition of MPE increased the particle size (based on the volume percentage) from 84.0 ± 4.4 µm in run 1 to 146.0 ± 5.0 µm in run 2. In contrast, the d (0.5) for MBR 1 (blank) decreased from 85.0 ± 4.0 µm in run 1 to 71.0 ± 3.0 µm at the end of run 2.

Figure 9 shows the particle number concentration (calculated based on the volume percentage distribution and sampling MLSS concentration in the MALVERN) in both reactors. The number concentration of particles in the MBR with MPE was significantly less than that of the MBR without MPE, but the particle size was larger. The d (0.5) based on the number percentage in MBR 1 and 2 were 1.55 and 9 µm respectively. These results suggest that the addition of MPE flocculated the small particles into larger particles, resulting in a decrease in the particle number concentration.

Figure 10 shows the viscosity of the mixed liquor in both reactors versus shear rate. From the figure, the viscosities of the non-flocculated and MPE flocculated sludges were similar over the shear rate range of 0–100 s⁻¹. Therefore, the difference in the biomass particle size distribution conferred by the 400 mg/L MPE dosage did not alter the rheological properties of the sludge significantly.

Figure 11 shows the average Specific Oxygen Uptake Rate (SOUR) of the blank and flocculated sludges. The SOUR of the sludge without MPE and flocculated sludge with MPE were 3.90 ± 0.12 mg/g/L and 3.75 ± 0.12 mg/g/L respectively. The SOUR for the flocculated sludge was slightly lower than the sludge without MPE and the difference in SOUR is around 4%. Although the flocculated sludge has a lower specific surface area due to its
larger particle size, the effective microbial activity of sludge with MPE was only slightly lower than that of the sludge without MPE. A possible explanation for this is that the viscosities of the sludges were similar and the 3–4 mg/L dissolved oxygen (DO) and substrate concentration in both MBRs were sufficiently high, so that the rate of oxygen and nutrient diffusion into the non-flocculated and flocculated sludge particle would be similar.

**Fouling Rate and Long Term Fouling of the Flocculated and Control MBR Activated Sludge in Run 2**

The flux stepping method was used to determine the apparent “critical flux.” The membranes applied in each flux step were cleaned to maintain constant clean water permeability. Figures 12a and b shows the TMP and flux against time for the flux-stepping experiments in run 2. The results are compared in Fig. 12c which shows that as the flux increased, the changing rate of the transmembrane pressure (dTMP/dt) increased. Above a flux of 20 LMH, the dTMP/dt for the MPE-modified sludge was always lower than the sludge of the control MBR. From Figure 12c, the fouling rate was almost zero below flux 20 LMH in control MBR and 30 LMH in the MBR with MPE, the fluxes represented the effective ‘critical fluxes’ in the MBRs. The addition of MPE led to a higher effective “critical flux.”

The long-term TMP profiles of control MBR and MBR with MPE were obtained at 3 different flux levels during run 2 and are shown in Fig. 13. A constant flux of 20 LMH which was approximately the “critical flux” of control the MBR and below the “critical flux” of the MBR with MPE was maintained in both MBRs. In MBR 1, the TMP increased gradually at an average rate of

<table>
<thead>
<tr>
<th>TABLE 3</th>
<th>Summary of microbial performance of MBR 1 &amp; 2 during the experiment</th>
</tr>
</thead>
<tbody>
<tr>
<td>Parameters</td>
<td>Startup period without adding MPE (run 1)</td>
</tr>
<tr>
<td>Parameters</td>
<td>MBR 1 (blank)</td>
</tr>
<tr>
<td>MLSS (g/L)</td>
<td>3.50–9.0 ± 0.73</td>
</tr>
<tr>
<td>Permeate TOC (mg/L)</td>
<td>3.50 ± 1.3</td>
</tr>
<tr>
<td>Total EPS (g/L)</td>
<td>0.40 ± 0.02</td>
</tr>
<tr>
<td>EPS/MLSS (mg/g)</td>
<td>40 ± 4</td>
</tr>
<tr>
<td>EPS in supernatant (mg/L)</td>
<td>18 ± 8–9.5 ± 2.1</td>
</tr>
<tr>
<td>TOC in supernatant (mg/L)</td>
<td>20 ± 5–7 ± 2.3</td>
</tr>
<tr>
<td>d0.5μm (particle size volume %)</td>
<td>85 ± 4</td>
</tr>
</tbody>
</table>
approximately 0.34 kPa/day for 27 days. In comparison, the \( \frac{dTMP}{dt} \) of MBR 2 increased at a slower rate of approximately 0.29 kPa/day after 27 days of operation.

A constant flux of 30 LMH was approximately the "critical flux" of MBR 2 (with MPE) and above the "critical flux" of the control MBR 1. It can be observed that the TMP of the membrane in MBR 2 was always lower than that of MBR 1. For the membrane in MBR 2, the TMP increased gradually at a rate of approximately 0.36 kPa/day for 19 days. The \( \frac{dTMP}{dt} \) of the membrane in MBR 1 increased at a faster rate of approximately 0.45 kPa/day after 14 days of operation, followed by a rapid increase in TMP (the "TMP jump" (3,4).

A flux level of 40 LMH was above the "critical flux" of the membranes in both MBRs with and without MPE. A rapid increase in TMP was observed for both reactors and the TMP reached 30 kPa after only 4 and 7 days of operation for MBR 1 and 2 respectively. However, the TMP of the membrane in MBR 2 was always lower than that of MBR 1.

FIG. 8. TMP and flux VS time during the start-up period (run 1).

FIG. 9. Particle size distribution of biomass with and without MPE.

FIG. 10. Viscosity of biomass with and without MPE.
In run 2, the flux stepping experiments showed that the effective "critical flux" increased after the sludge had been modified by MPE and the results of the long-term experiments confirmed lower membrane fouling after the addition of MPE. A comparison of the performances of the two MBRs shows that the lower fouling in MBR 2 could be due to the larger flocculated particles. At the "critical flux" of 20 LMH and 30 LMH for MBR 1 and 2 respectively, the fouling rate was similar at 0.34 kPa/day and 0.36 kPa/day respectively. This agrees with the pilot MBR data of Soong et al. (19), who observed a 50% increase in the operating flux with MPE addition. In addition, the membrane performances of both MBRs were better than in run 1, probably due to the low level of supernatant EPS and TOC in run 2.

Termination of MPE Addition in MBR 2 (Run 3)

Biomass Characteristics in Run 3

In run 2, the daily dosage of MPE to maintain a level of about 400 mg/L was continued until MBR 2 reached steady state. From the 122nd to the 155th day (run 3), the addition of maintenance MPE was terminated in MBR 2 to study the effect of discontinued MPE addition on membrane fouling. It should be noted that from the 110th – 122nd day at the end of run 2, the conditions in both reactors were somewhat disturbed due to the frequent membrane cleaning for the flux stepping experiments. The fluctuations in the bioreactors can be observed in Fig. 6, where the supernatant TOC in both MBR 1 and 2 increased. However, in run 3 MBR 1 reached a new steady state while MBR 2 (terminated MPE) continued to increase.
in a fluctuating pattern. The concentrations of MLSS, total EPS and EPS/MLSS, EPS in the supernatant, TOC of the permeate, and TOC of the supernatant and particle size in this period are shown in Fig. 2–7 respectively.

The MLSS concentrations in MBR 1 and 2 were relatively stable at $10.20 \pm 0.56$ g/L and $9.90 \pm 0.34$ g/L respectively in run 3 as observed in Fig. 2. The termination of MPE addition in MBR 2 did not affect the MLSS concentration and the MLSS concentrations in both reactors were relatively constant during this period.

In Fig. 5, the average permeate TOC concentrations of MBR 1 and 2 were $2.1 \pm 0.3$ mg/L and $2.4 \pm 0.2$ mg/L respectively, indicating a similar organic removal in run 3. However, as shown in Fig. 6, terminating MPE addition in MBR 2 increased the supernatant TOC of the mixed liquor dramatically from $6.7 \pm 0.2$ mg/L in run 2 to $70.0 \pm 6.0$ mg/L in run 3 while the supernatant TOC of MBR 1 remained at about $20.0$ mg/L.

In Fig. 3, the total EPS in MBR 2 increased from $0.40$ g/L to $0.67$ g/L during run 3 while the total EPS in MBR 1 stabilized at around $0.40$ g/L after a small peak. The average EPS/MLSS was $31.0 \pm 6.0$ mg/gMLSS and $51.0 \pm 6.0$ mg/gMLSS in MBR 1 and 2 respectively. The supernatant EPS of MBR 1 and 2 showed an apparent difference in run 3 (Fig. 4). The average supernatant EPS remained at $20.0 \pm 3.0$ mg/L in MBR 1 but increased from $7.8 \pm 4.5$ mg/L to $62.0 \pm 9.0$ mg/L in MBR 2. The EPS analysis showed that the total EPS, EPS/MLSS, and EPS in the supernatant increased in MBR 2 after the termination of the maintenance MPE dosage.

In Fig. 7, the particle size (based on volume percentage) remains constant in MBR 1, while in MBR 2 the particle size based on the volume percentage decreased after the MPE addition was stopped. This result suggests that the flocculated particles disintegrated as the level of MPE dropped in the reactor. Figure 9 shows the particle number concentration in MBR 2 for runs 2 and 3. The particle number concentration in MBR 2 showed a higher peak and shifted to the left, indicating an increased amount of smaller particles after the termination of the maintenance MPE dosage.

![FIG. 13. TMP vs time at flux 20, 30, 40 LMH in long term operation in MBR with and without MPE.](image)

![FIG. 14. TMP vs time at flux 20 LMH after the termination of MPE addition in MBR 2.](image)
CONCLUSIONS

The effect of MPE addition on the performance of the submerged membrane bioreactor was investigated in terms of the TMP increase of the membrane under constant flux operation and the following conclusions can be drawn:

1. In run 1 (start-up), similar biomass characteristics and fouling intensity were achieved in the parallel MBRs after 60 days of operation.
2. In run 2 (MPE to MBR 2), the supernatant EPS and SMP were maintained at the same low level in both reactors. After adding MPE to MBR 2, the flocculation of the activated sludge by MPE resulted in larger particle size distribution, mitigating the membrane fouling.
3. The addition of the MPE flocculant resulted in a 50% increase in “critical flux.” However, below the effective “critical flux,” there was negligible difference in terms of fouling in the 2 parallel MBRs.
4. In run 3, the termination of the MPE maintenance dosage resulted in an increase in smaller particles, supernatant EPS, and TOC in MBR 2, leading to more severe membrane fouling than the control MBR. Continuous addition of MPE is recommended once the flocculant is applied.

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