Membrane fouling and performance evaluation of conventional membrane bioreactor (MBR), moving biofilm MBR and oxic/anoxic MBR

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ABSTRACT

In this study, three laboratory scale submerged membrane bioreactors (MBRs) comprising a conventional MBR (C-MBR), moving bed MBR (MB-MBR) and anoxic-oxic MBR (A/O-MBR) were continuously operated with synthesized domestic wastewater (chemical oxygen demand, COD = 500 mg/L) for 150 days under similar operational and environmental conditions. Kaldnes® plastic media with 20% dry volume was used as a biofilm carrier in the MB-MBR and A/O-MBR. The treatment performance and fouling propensity of the MBRs were evaluated. The effect of cake layer formation in all three MBRs was almost the same. However, pore blocking caused a major difference in the resultant water flux. The A/O-MBR showed the highest total nitrogen and phosphorus (PO₄-P) removal efficiencies of 83.2 and 69.7%, respectively. Due to the high removal of nitrogen, fewer protein contents were found in the soluble and bound extracellular polymeric substances (EPS) of the A/O-MBR. Fouling trends of the MBRs showed 12, 14 and 20 days filtration cycles for C-MBR, MB-MBR and A/O-MBR, respectively. A 25% reduction of the soluble EPS and a 37% reduction of the bound EPS concentrations in A/O-MBR compared with C-MBR was a major contributing factor for fouling retardation and the enhanced filtration capacity of the A/O-MBR.

Key words | anoxic zone, extracellular polymeric substances (EPS), membrane fouling, nitrogen removal, plastic media, simultaneous nitrification and denitrification (SND)

INTRODUCTION

Disposal of untreated wastewater from residential or industrial areas can result in adverse soil pollution and surface water contamination (Tanner et al. 2012) requiring adequate treatment to satisfy local discharge standards for surface water bodies. The membrane bioreactor (MBR) is an efficient technology for treating domestic and industrial wastewater to a very high quality. The MBR is a combination of a biological process and a membrane filtration process, the latter using either micro or ultra-filtration (Kim et al. 2011). The MBR enjoys many advantages over the conventional activated sludge process for wastewater treatment, in terms of both treatment efficiency and process control (Miura et al. 2007). Despite all the advantages of MBR, it is still not being implemented at the level it might be. The main reason for this is the high energy and chemical cost for membrane fouling control (Huyskens et al. 2012).

It is generally accepted that fouling reduces the performance of a membrane. When fouling occurs, a thick gel layer or cake layer is formed on the membrane surface, causing the permeate flux to decline. Fouling is usually attributed to a number of parameters, such as sludge particle deposition, adhesion of macromolecules such as extracellular polymeric substances (EPS), and pore clogging by small molecules and/or colloids. The nature of the fouling is strongly influenced by biomass characteristics, operating conditions and membrane characteristics (Sweity et al. 2011).

EPS are the metabolic products of active bacterial secretion, and can be in either soluble or bound form. They consist of proteins, polysaccharides, nucleic acids and lipids accumulating on the bacterial cell surface. EPS are the main reason for the adhesion of sludge flocs to the membrane, and are recognized as the most significant...
factor affecting biofouling in MBRs (Laspidou & Rittmann 2002). EPS production and accumulation on the membrane are influenced by several factors. Many studies have linked EPS with the solid retention time (SRT) of the MBR (Clech et al. 2006). At shorter SRT, sludge is found to produce more EPS compared with longer SRT sludge (Cho et al. 2005). Increasing the SRT can support the development of slowly growing microorganisms that are able to consume polysaccharides and proteins as substrates, and produce less biopolymers (Masse et al. 2006). Ouyang & Liu (2009) reported higher soluble EPS concentrations at shorter SRT, with protein concentrations higher than polysaccharides. But one needs to compromise between a high fouling tendency at short SRT and a high viscosity of mixed liquor at very long SRT (Ouyang & Liu 2009).

Membrane fouling can be limited by either operating MBRs at low flux, that is, below critical flux or by increasing air scouring rate. Physical relaxation, intermittent filtration and backwashing are good methods to reduce membrane fouling (Wang et al. 2008). In the moving bed-MBR (MB-MBR), the biomass grows on carriers that move freely in the MBR by aeration or by mechanical mixing. MB-MBR systems experience enhanced treatment capabilities and minimized fouling by allowing the biomass growth in attached form over support carriers (Hu et al. 2012). Mechanical mixing along with diffused aeration keeps the sludge simultaneously in anoxic and oxic conditions in an anoxic/oxic-MBR (A/O-MBR). Such an A/O-MBR is found to be an appropriate solution for membrane fouling mitigation (Pradhan et al. 2012).

There are various reactor configurations and modifications available for membrane fouling mitigation in MBR systems to prolong the filtration cycle without disturbing treatment efficiency. In this study, an effort has been made to compare treatment efficiencies and fouling tendencies in three differently configured MBRs, namely the conventional MBR (C-MBR), the MB-MBR and the A/O-MBR. These MBRs were operated for 150 days in parallel under similar operating and environmental conditions. The treatment performance and membrane fouling properties were investigated, and the factors affecting membrane fouling were identified.

**METHODS**

**Experimental setup**

Three laboratory scale MBRs: C-MBR, MB-MBR and A/O-MBR made of acrylic (transparent plastic) were operated in parallel at steady state for 150 days. The reactors were rectangular in shape with working volumes of 12, 12 and 16 L for C-MBR, MB-MBR and A/O-MBR, respectively. Prior to the steady state condition, 2 months were allowed for acclimatization of seed sludge obtained from the sewage treatment plant, Sector 1-9, Islamabad, Pakistan. Details of the experimental setup for all three MBRs are shown in Figure 1. Hollow fiber (HF) membranes made up of polyvinylidene fluoride (PVDF) with a pore size of 0.1 μm and an effective surface area of 0.2 m² (Mitsubishi Rayon, Japan) were used in all three MBRs in submerged mode. A peristaltic pump (Master Flex, Cole-Parmer, USA) operating in a 10 min filtration and 2 min relaxation cycle was used to extract the treated water from HF modules under suction pressure. Aeration at the rate of 5 L/min was provided continuously through fine bubble air diffusers to keep the sludge in suspension and to maintain the required dissolved oxygen (DO) provision (2–5 mg/L). A data logging manometer (Sper-Scientific 840099, Taiwan) connected to the membrane suction line was applied for continuous data recording of the trans-membrane pressure (TMP). The membranes were operated up to a maximum TMP of 30 kPa and then physico-chemical cleaning was applied. The protocols for maximum operating TMP and membrane cleaning were provided by the manufacturer (Mitsubishi Rayon, Japan). Kaldnes® plastic media with a dry volume of 20% and a wet volume of 8% was used in the study. The properties of the media and the composition of the synthetic domestic wastewater are described in the previous work (Khan et al. 2013). The C-MBR was operated as a control, with suspended biomass only. In the MB-MBR, the Kaldnes® plastic media was added as a moving carrier for supporting the attached growth. In the A/O MBR, an anoxic zone was created in one compartment by utilizing mechanical mixing along with Kaldnes® plastic media. The mechanical mixer in the anoxic zone kept the sludge and media in suspension and managed to maintain a DO level between 0.5–1.0 mg/L, as compared with a DO between 2–4 mg/L in the oxic (aerobic) zone utilizing diffused aeration.

**Operating conditions**

The MBRs were operated at an 8 h hydraulic retention time (HRT) and 30 days SRT. The organic loading rate and nitrogen loading rate were maintained at 1.5 kg-COD/m³/d and 0.15 kg-NH₄-N/m³/d, respectively in all three MBRs, while the food/microorganism ratio (F/M) was maintained at 0.25, 0.18 and 0.22/day in the C-MBR, MB-MBR and A/O MBR, respectively.
Analytical methods

All parameters mentioned in Table 1 were measured as per Standard Methods (APHA 2005). Specific oxygen uptake rate (SOUR) was measured to determine microbial activity via dissolved oxygen (DO) depletion rate in pre-aerated sludge (Xing et al. 2001). Soluble EPS was analyzed by centrifugation of a sludge sample. Bound EPS was extracted by the cation exchange resin method (Frolund et al. 1996). Carbohydrate and protein concentrations were measured by using a spectrophotometer, adopting the methods employed by Dubois and Lowry (Lowry et al. 1951; Dubois...
Particle size distribution (PSD) and the mean sludge particle size in the MBRs were measured using the light scattering technique. The specific cake resistance (SCR) and membrane resistances were measured according to the method and formulas used in a previous study (Khan et al. 2015).

RESULTS AND DISCUSSION

Performance evaluation of the MBRs

Table 1 summarizes the results of the treatment performance of the three MBRs. The reported values include the average and standard deviation over 90 days.

The MB-MBR showed a higher mixed liquor suspended solids (MLSS) concentration as compared to the C-MBR. This may be attributed to the bio-support plastic media, which provided space for the accumulation of bacterial biofilms. The reduced MLSS concentration in the A/O MBR compared with the MB-MBR may be attributed to the anoxic conditions in the A/O-MBR, where the combination of slow growing nitrifying and de-nitrifying bacteria could be the predominant microbial species. The organic removal efficiency remained above 95%, with minimal variation among the three MBRs. The A/O-MBR exhibited substantially higher total nitrogen (TN) and phosphate-phosphorus (PO₄-P) removal efficiencies; that is, about 23% greater than the C-MBR. It may be linked to effective simultaneous nitrification and denitrification (SND) in the A/O-MBR, due to the combined effect of the anoxic and oxic zones, which has been proven in the past to be a good strategy for nitrogen removal (Kimura et al. 2008). Denitrification has been linked to enhanced phosphorus uptake by the denitrifying phosphorus accumulating microorganisms and their subsequent removal as excess sludge (Parco et al. 2009).

Membrane fouling tendencies

Membrane fouling tendencies during the study were determined by continuously recording the TMP. Figure 2 shows the four consecutive TMP profile cycles of all the three MBRs. On average, the C-MBR fouled after 12 days, the MB-MBR after 14 days and the A/O-MBR after 20 days. Use of Kaldnes® media enhanced the membrane filtration cycle in both the MB-MBR and the A/O-MBR compared with the C-MBR. Kaldnes® media played an important role in reduction of the cake layer over the membrane surface due to the scouring of the membrane. The introduction of an anoxic zone in the A/O MBR further reduced the fouling propensity by prolonging the filtration runs by almost twice compared to that in the C-MBR.

PSD effect on membrane fouling

The PSD of all three MBRs is shown in Figure 3. The C-MBR showed a broader distribution but the use of Kaldnes® media caused a narrowing of the distribution curve in the MB-MBR and the A/O-MBR by breaking the flocs into smaller sizes. The breakage occurred because of the physical collision of the flocs with the edges of the biofilm carriers. The mean particle size was 112 ± 24, 33 ± 8, and 41 ± 9 µm in the C-MBR, MB-MBR and A/O MBR, respectively.

![Figure 2](https://iwaponline.com/wst/article-pdf/69/7/1403/472335/1403.pdf)

**Table 1 | Summary of the performance evaluation of the MBRs**

<table>
<thead>
<tr>
<th>Analysis</th>
<th>C-MBR</th>
<th>MB-MBR</th>
<th>A/O-MBR</th>
</tr>
</thead>
<tbody>
<tr>
<td>MLSS (g/L)</td>
<td>6.2 ± 1.04</td>
<td>8.2 ± 0.72</td>
<td>6.6 ± 0.65</td>
</tr>
<tr>
<td>MLVSS (g/L)</td>
<td>5.0 ± 0.83</td>
<td>6.4 ± 0.67</td>
<td>5.3 ± 0.70</td>
</tr>
<tr>
<td>Effluent COD (mg/L)</td>
<td>25.5 ± 12.3</td>
<td>20.9 ± 12.1</td>
<td>19 ± 11.3</td>
</tr>
<tr>
<td>COD removal %</td>
<td>95.0 ± 2.3</td>
<td>95.8 ± 2.5</td>
<td>96.3 ± 2.4</td>
</tr>
<tr>
<td>NH₄-N (mg/L)</td>
<td>5.7 ± 0.6</td>
<td>2.6 ± 0.6</td>
<td>2.2 ± 0.4</td>
</tr>
<tr>
<td>NO₂-N (mg/L)</td>
<td>2.5 ± 0.8</td>
<td>2.7 ± 0.7</td>
<td>1.1 ± 0.2</td>
</tr>
<tr>
<td>NO₃-N (mg/L)</td>
<td>12.1 ± 1.8</td>
<td>10.5 ± 1.5</td>
<td>5.2 ± 0.7</td>
</tr>
<tr>
<td>TN (mg/L)</td>
<td>20.3 ± 1.7</td>
<td>15.7 ± 1.4</td>
<td>8.5 ± 0.7</td>
</tr>
<tr>
<td>TN % removal</td>
<td>59.8 ± 3.6</td>
<td>68.8 ± 3.0</td>
<td>83.2 ± 1.7</td>
</tr>
<tr>
<td>Effluent PO₄-P (mg/L)</td>
<td>7.6 ± 1.5</td>
<td>5.8 ± 1.2</td>
<td>4.3 ± 1.4</td>
</tr>
<tr>
<td>PO₄-P % removal</td>
<td>46.5 ± 6.7</td>
<td>59.5 ± 4.4</td>
<td>69.7 ± 7.7</td>
</tr>
</tbody>
</table>

MLSS: mixed liquor suspended solids; MLVSS: mixed liquor volatile suspended solids; COD: chemical oxygen demand.
In all the MBRs, the sludge particle size was much larger than the pore size of the membrane, so it may be assumed that the bio-floc morphology is not a major contributing factor to fouling tendencies in the MBRs. An increase in the particle size of the A/O-MBR compared with the MB-MBR suggests that mechanical mixing causes less floc breakage compared with the shear force exerted by diffused aeration.

**SCR effect on membrane fouling**

The SCR of MBR sludge was measured by dead end filtration setup (Amicon, Model 8400, USA) and the SCR of C-MBR, MB-MBR and A/O-MBR were found to be $4.72 \times 10^{16}$, $2.51 \times 10^{15}$ and $1.25 \times 10^{15}$ m/kg, respectively. SCR gives a measure of fouling potential of the MBR biomass; with high values of SCR, the fouling propensity of such an MBR system should increase rapidly (Park et al. 2006). The significant difference in SCR values between the C-MBR and the other two MBRs is due to the presence of Kaldnes® media which serves as a biofilm carrier and reduces the sludge load over the membrane surface (Yang & Zhang 2006). This implies that the intensity of cake layer formation over membrane fibers in both the MB-MBR and the A/O-MBR should be less than the C-MBR.

**SOUR effect on membrane fouling**

The SOUR was measured in terms of carbonaceous and nitrogenous uptake rates with substrates rich in carbon and nitrogen, respectively. The carbonaceous SOUR was found to be 179.4, 186.8 and 167.8 mg-O$_2$/g-VSS/h while the nitrogenous SOUR was found to be 26.9, 19.0 and 21.7 mg-O$_2$/g-VSS/h in the C-MBR, MB-MBR and A/O-MBR, respectively. Higher MLSS and carbonaceous SOUR in the MB-MBR indicates higher microbial activity compared with the other MBRs. However, biofilm formation over moving carriers reduced the sludge load and resultant cake formation on the membrane surface. Hence, the MB-MBR fouled later than the C-MBR. The A/O-MBR had comparatively lower carbonaceous SOUR than the MB-MBR, resulting in a lower biomass concentration and thus was further facilitated to improve the filtration cycle. In this study, the impact of the relatively small nitrogenous SOUR variation among the MBRs was found to have an insignificant impact on fouling tendencies. In the absence of an anoxic chamber and media (having a partial anoxic environment) in C-MBR, favorable conditions in terms of uniform high DO (4–5 mg/L) for growth of nitrifiers (ammonium oxidizing bacteria) was available, resulting in high nitrogenous SOUR as well as a high effluent NO$_3$-N concentration in C-MBR compared with the other two MBR systems.

**EPS effect on membrane fouling**

Table 2 summarizes the extracellular polymeric substance (EPS) concentrations in the three MBRs.

The C-MBR showed the highest soluble and bound EPS concentrations followed by the MB-MBR and finally the A/O-MBR. The soluble EPS concentration was reduced by 17 and 25% in the MB-MBR and A/O-MBR, respectively, while the bound EPS concentration was reduced by 22 and 37% in the MB-MBR and A/O-MBR, respectively, compared with that in the C-MBR. These results reveal that the influence of the A/O-MBR configuration and the presence of moving plastic carriers on soluble and bound EPS were significant. The protein content showed a major contribution of 62–66% of the soluble EPS and 66–83% of the bound EPS among the MBRs while the remaining contribution was associated with carbohydrate content. Proteins are macro-molecules consisting of many amino acids, where nitrogen is the basic building and binding element. Proteins can be bent in any direction to form 3-D structures. These structures may also trap other flocs in the sludge and get deposited on the membrane surface in the MBR. Since the highest TN removal of 83% was achieved in the A/O-MBR, the protein levels consequently dropped substantially compared with the other two MBRs. This fact reveals that the significant decrease in nitrogen concentration in the A/O-MBR was the main reason for the decrease in soluble and bound EPS protein concentrations, which can be considered as the major...
factor responsible for membrane fouling retardation. However, the effects of decline in soluble and bound carbohydrate contents cannot be ignored in terms of a prolonged filtration cycle in A/O-MBR. The overall lowest EPS concentrations, as found in the A/O-MBR, may be responsible for a retarded fouling behavior and an enhanced filtration capability.

Membrane resistance analysis

A summary of the resistance analysis of all three MBRs is presented in Table 3. The decrease in total hydraulic resistance ($R_t$) of the MB-MBR and A/O-MBR compared with the C-MBR was 17 and 27%, respectively. The difference in $R_t$ of the three MBRs was due to the decrease in cake layer resistance ($R_c$) as well as pore blocking resistance ($R_p$). Furthermore, 22 and 57% reduction of $R_p$ was observed in the MB-MBR and A/O-MBR compared with the C-MBR, respectively while $R_c$ did not vary significantly in case of A/O-MBR. The significant decrease in $R_t$ in the A/O-MBR can be attributed to the lower soluble and bound EPS concentrations due to the reduction in protein content. The decline in soluble EPS concentration of 25% in A/O-MBR compared with C-MBR was almost similar to the decrease in the $R_t$ value of 27% which implies that soluble EPS concentration has a major influence on the $R_t$.

**CONCLUSIONS**

This study investigated the fouling behavior of the C-MBR, MB-MBR and A/O-MBR under similar operating conditions. Results revealed that the introduction of the anoxic zone in the A/O-MBR augmented the total nitrogen removal efficiency by 23% as compared with the C-MBR. Nitrogen is the key element in joining many amino acid units through peptide linkage to form proteins. Thus, in the A/O-MBR the soluble and bound EPS contents were reduced by 25 and 37%, respectively compared with the C-MBR. Membrane resistance analysis demonstrated that the major contributor to the fouling in the C-MBR was pore blocking resistance ($R_p$). A decrease by 57% of $R_p$ in the A/O-MBR resulted in a 67% long filtration cycle of 20 days compared with a 12 day cycle in the C-MBR. Therefore, a combination of anoxic and oxic zone compartments and the introduction of biofilm growth over moving plastic carriers were favorable in MBR operation, in terms of both effective organic and nutrient removal and membrane fouling retardation.
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