EFFECTS OF FINE PARTICLES ON PERFORMANCE OF ENHANCED BACKWASH FOR FOULING REDUCTION IN ANAEROBIC MEMBRANE BIOREACTORS

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EXTENDED ABSTRACT

Anaerobic membrane bioreactor (AnMBR) can produce high-quality effluents through effective solid-liquid separation using microfiltration. During the operation of the membrane bioreactor system, membrane fouling is one of the main problems. Several models have been proposed to understand the mechanisms of fouling of the membrane. Usually, cake formation and pore blocking are found to be the main mechanisms. Many factors such as hydrodynamic conditions, membrane materials, and sludge properties are involved in fouling. Particle size distributions and extracellular polymeric substance concentrations in sludge properties have been suggested as being key parameters in determining the degree of fouling in an AnMBR. To control membrane fouling, numerous methods are used during operation, such as bubbling of biogas produced in the AnMBR, backwash with permeate, and enhanced backwash, in which a chemical reagent is introduced to dissolve foulants adsorbed to the membrane surfaces. Enhanced backwash is effective in recovering flux from high-strength organic feed solutions; however, little research has been conducted on the effects of sludge properties on the efficiency of enhanced backwash in an AnMBR.

The objectives of this research are to investigate effects of sludge properties in terms of particle composition on performance of enhanced backwash and to understand the fouling mechanisms of specific feed solutions with different particle compositions. The sludge was centrifuged at three rotational speeds to differentiate particle size distributions, and the supernatant and residual flocs were used for microfiltration and enhanced-backwash process. The filtration flux with the flocs was greater than those of the three supernatants, which indicates that fouling was greater with supernatants. Taking into account that the solid concentrations of the raw sludge and the residual flocs were 10 times greater than those of the supernatants, the reduction in flux for the supernatants is significant. The flux for the solution centrifuged at 500 rpm was the lowest and those of the two solutions treated at 1000 and 1500 rpm were similar, which implies that certain ranges of particle size are particularly important for fouling in the AnMBR. The flux recovery was found to be approximately 2225 L/m²/h by physical cleaning and 2743 L/m²/h by NaOCl cleaning. The efficiency of enhanced backwashing was also lowest for the solution centrifuged at 500 rpm. Particle composition such as the fine-particle concentration played a significant role in fouling and the chemical cleaning efficiency.

Keywords: Anaerobic MBR; Membrane fouling; Supernatant; Enhanced backwash; PTFE microfilter
1. INTRODUCTION

Owing to the possibility of energy generation and the efficient separation of two sludge phases, anaerobic membrane bioreactors (AnMBRs) have been received significant attention in wastewater treatment (Lin et al., 2013). Compared to aerobic treatments, bio-energy gases such as methane (CH\textsubscript{4}) can be generated without aeration (Skouteris et al., 2012). According to Hu and Stuckey (2006), the AnMBR not only allows short hydraulic retention times (HRTs) independent to solids retention times (SRTs), but also can improve effluent water quality and nutrient (nitrogen and phosphorus) recycling potential as a fertilizer. AnMBR has been proven to provide more reliable and more compact anaerobic treatment compared to traditional anaerobic processes.

The critical bottleneck of the AnMBR process is membrane fouling, which limits the operability of the process. Membrane fouling can decrease the productivity and increase the cost of the system (Lin et al., 2013), and controlling membrane fouling would accelerate membrane applications. The mechanism of membrane fouling has been categorized in many studies using Hermia’s model. There are four fouling mechanisms: pore blocking, complete blocking, intermediate blocking, and cake layer formation. According to the Hermia’s model, the major membrane fouling mechanisms under constant pressure filtration were found to be cake formation or complete blocking (Liu et al., 2012; Lin et al., 2013; Meng et al., 2009). According to Meng et al. (2009), there are numerous factors that affect membrane fouling, such as hydrodynamic conditions, membrane materials, module design, and sludge properties including particle size, extracellular polymeric substances (EPS), hydrophobicity, and surface charge. In particular, particle size is one of the key factors that controls membrane fouling permeability (Lin et al., 2009; Liu, 2009; Marco, 1997). Liu (2009) reported that particle sizes of 1.0~2.7 µm mainly contributed to 87% of cake resistance, assuming cake formation and pore blocking. Marco (1997) showed that increasing particle size through coagulation and sedimentation results in a higher permeability and reduces fouling.

There are several ways to control fouling in membrane processes, such as development of anti-fouling membranes, use of good-quality feed water, and optimization of operations to reduce fouling. Stuckey (2012) categorized the fouling control strategies during operation into three approaches: (1) scheduled continuous cleaning by both relaxing/backflushing and chemical cleaning while maintaining a high flux, (2) less chemical cleaning than in the first approach and operating below a critical flux, and (3) using efficient operational procedures such as employing hydrodynamic tools or adsorbent additions to the AnMBR reactor. The most frequently applied practice for fouling control in both MBRs and AnMBRs is periodic backwash with permeate (Kim et al., 2001, Lew et al., 2009, Zhang et al., 2007). Backwash can alleviate fouling, but for only a short period of time. Compared to regular backwash, enhanced backwash has been proposed as a more efficient backwash method and is popular in membrane processes in water and wastewater treatment plants. During enhanced backwash, low doses of one or a mixture of chemical agents such as NaOCl and NaOH are intermittently introduced. For example, operations could consist of the usual filtration, regular backwash every 30 minutes, and enhanced backwash every 12 hours. Enhanced backwash is effective for flux recovery in high-strength organic feed solutions. However, little research has been conducted on the effect of sludge properties on enhanced-backwash efficiency in AnMBRs.

Thus, in this study, sludge properties were differed by particle compositions, more specifically particle size distributions. The sludge was centrifuged at three rotational speeds to yield different particle size distributions. The supernatant, the residual flocs, and the raw sludge were used for microfiltration and the enhanced-backwash process. Results from the three types of feed waters were evaluated for performance of enhanced backwash and for better understanding of fouling mechanisms.
2. THEORETICAL MODELS

Resistance-in-series model
A flux model is useful for obtaining membrane operating parameters such as membrane resistances. The resistance-in-series model has been used to understand flux and fouling characteristics in microfiltration. The permeate flux can be expressed in terms of resistances and the transmembrane pressure (Wiesner et al., 1996). Cake resistance and specific cake resistances can be obtained from the resistance-in-series model evaluated using flux measurements for a solution. In addition, resistances incorporate the operational process of microfiltration.

\[
J = \frac{\Delta P}{\mu \cdot R_T} = \frac{\Delta P}{\mu \cdot (R_M + R_{ph} + R_{ch} + R_{ir})}
\]

where \( J \) is the permeate flux through the membrane; \( \Delta P \), the transmembrane pressure; \( \mu \), the viscosity of permeate; \( R_T \), the total resistance; \( R_M \), the intrinsic membrane resistance; \( R_{ph} \), the cake resistance, which can be recovered by physical cleaning; \( R_{ch} \), the foulant resistance, which can be recovered by chemically enhanced-backwash cleaning; and \( R_{ir} \) the irreversible fouling resistance, which is not recovered by the cleaning procedures applied in this study. The clean-water flux of the new membrane was used to obtain \( R_M \). The flux at the end of the sludge filtration, after physical cleaning, and after NaOCl cleaning were used to calculate \( R_T \), \( R_{ph} \), and \( R_{ch} \), respectively. The irreversible fouling resistance was calculated from the difference between the total resistance and the sum of the resistances recovered by physical and chemical cleaning.

3. EXPERIMENTAL

3.1. Raw sludge and feed solutions

Primary sludge was taken from an anaerobic digester (AD) in the Ansan wastewater treatment plant (Gyunggi Province, Korea). The AD was operated at a temperature of 35°C and a hydraulic retention time of 39 days. The primary sludge from the digester, which had a total solid concentration of approximately 23900 mg/L, was sampled and moved to the laboratory for membrane filtration tests on that day, so that the sludge activity was maintained at the same level as in real operations. The sludge was diluted to a concentration of 3000 mg TS/L for further experiments on microfiltration and cleaning efficiency. The raw sludge with 3000 mg SS/L was then centrifuged for 10mins (VS-6000N, Vision Scientific Co., Korea) at three rotational speeds, i.e., 500, 1000 and 1500 rpm, to produce solutions with different particle properties such as particle size distribution. Centrifugal sedimentation is used to separate submicron particles (Gee, 2002). A particle in a centrifugal field settles with a velocity which established by tow forces in opposition, a centrifugal force and a drag force. The settling velocity from centrifugal sedimentation is increased proportional to the square of particle diameters and of rotational speeds. The different rotational speeds therefore yield supernatants with sizes of particle that are not settled under the condition (Foster, 1992; Allen, 1996). The bigger particles settle at the smaller rotational speed. From Stokes’ equation (Gee, 2002), the average settled particle sizes showed the tendency with 28.96, 14.48 and 9.65µm at each speeds. The fine particle distributions of supernatants were estimated less than each average settled particle sizes. After the supernatants were removed, the remaining flocs were completely mixed with distilled water of the same volume as that of the liquid withdrawn. The supernatants from centrifugation and the remaining flocs were used for microfiltration and the enhanced-backwash process. Characteristics of the key water quality parameters of the feed waters are shown in Table 1.
Table 1. Characteristics of feed solution before microfiltration

<table>
<thead>
<tr>
<th>Parameters</th>
<th>SS(mg/L)</th>
<th>Turbidity (NTU)</th>
<th>pH</th>
<th>UV254 (cm⁻¹)</th>
<th>COD(mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raw sludge</td>
<td>3000</td>
<td>580</td>
<td>7.79</td>
<td>0.256</td>
<td>116</td>
</tr>
<tr>
<td>Supernatants</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>SC500 from centrifuge*</td>
<td>-</td>
<td>63.9</td>
<td>7.93</td>
<td>0.241</td>
<td>26</td>
</tr>
<tr>
<td>SC1000</td>
<td></td>
<td>46.7</td>
<td>7.68</td>
<td>0.135</td>
<td>8</td>
</tr>
<tr>
<td>SC1500</td>
<td></td>
<td>38.2</td>
<td>7.82</td>
<td>0.197</td>
<td>4</td>
</tr>
<tr>
<td>Flocs</td>
<td>19067</td>
<td>343</td>
<td>6.77</td>
<td>0.018</td>
<td>-</td>
</tr>
</tbody>
</table>

*SC500, SC1000, and SC1500 are supernatants centrifuged at a speed of 500 rpm, 1000 rpm, and 1500 rpm, respectively.

3.2. Membrane filtration

A bench-scale membrane apparatus (Millipore Co. USA) was set up to conduct short-term filtration tests for different concentrations of the enhanced-backwash agents. A schematic of the bench-scale membrane apparatus is shown in Figure 1. The feeds for microfiltration were introduced into a 3-L reservoir and then moved to the batch cell for dead-end operation by nitrogen gas. The nitrogen gas was used to maintain a constant pressure of 14.5 psi (i.e., 100 kPa). Permeate from the cell was measured with an electronic balance (AND GF-2000, A&D Engineering Inc., San Jose, USA), which was connected to a data acquisition system to record the mass of water every 20 s.

![Figure 1. Schematic of bench-scale membrane setup](image)

The PTFE used in the batch cell had an effective area of 28.7 cm² with further properties listed in Table 2. The clean-water flux of each filter was measured for 10 minutes after one day of soaking in distilled/deionized water. The washing includes 10 minutes of physical cleaning and 10 minutes of chemical cleaning with 100 mL of NaOCl. The enhanced-backwash reagent was added after physical cleaning with a stirring bar. The clean-water flux with distilled water (PURELAB classic, ELGA LabWater, Lane end, UK) was measured after the each cleaning procedure.

Table 2. Properties of PTFE membrane filter used in this study

<table>
<thead>
<tr>
<th>Applications</th>
<th>Pore Size (µm)</th>
<th>Thickness (µm)</th>
<th>Flow Time* (mL/min/cm²)</th>
<th>Porosity (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hydrophilic PTFE</td>
<td>0.1</td>
<td>30</td>
<td>100</td>
<td>80</td>
</tr>
</tbody>
</table>

*100-mL water, 20 °C, 47-mm disk, 8.97”-Hg vacuum conditions were used (provided by the manufacturer)
3.3. Analytical methods

Chlorine solutions with a concentration of 100 mg/L were formulated from 8% NaOCl stock solution (DukSan, Seoul, Korea). The chlorine concentration was standardized by titrating the solution with 0.01 N Na\textsubscript{2}S\textsubscript{2}O\textsubscript{3} followed by using Standard Methods (20\textsuperscript{th}ed., APHA, AWWA, and WEF, 1998). The dose specified for the NaOCl cleaning was then diluted from the stock solution. The total solids were also measured using 10 mL of the sludge via the Standard Method. A Hach 2100N turbidimeter (Hach, Loveland, CO, USA) was used to measure turbidity and a StablCal\textsuperscript{®} Calibration Set was used for calibration. pH was measured using an Orion Model 410+ApH meter (Thermo Electron Co., Beverly, MA, USA). Chemical oxygen demand was measured using a reactor digestion method with a UV-visible spectrophotometer (DR2500, Hach Co., USA). COD measurements were taken after filtering the samples through the pre-rinsed 0.45-μm membrane syringe filters.

4. RESULTS AND DISCUSSION

4.1. Microfiltration performance

The diluted anaerobic digester sludge and solutions treated at different centrifuge speeds underwent microfiltration. The water quality of the microfiltration permeate was relatively high in terms of turbidity and UV\textsubscript{254} absorbance removal, as shown in Table 3. The turbidity of the permeate decreased to less than 0.51 NTU for the entire experiment. Organic removal, which was measured using the UV\textsubscript{254} absorbance, was in the range of 36.5–66.7%, which is substantial in an AnMBR application.

Table 3. Water quality of microfiltration permeates

<table>
<thead>
<tr>
<th>Permeate</th>
<th>Raw sludge</th>
<th>SC500</th>
<th>SC1000</th>
<th>SC1500</th>
<th>Flocs</th>
</tr>
</thead>
<tbody>
<tr>
<td>Turbidity (NTU)</td>
<td>0.389</td>
<td>0.228</td>
<td>0.511</td>
<td>0.367</td>
<td>0.128</td>
</tr>
<tr>
<td>% removal</td>
<td>99.9</td>
<td>99.6</td>
<td>98.9</td>
<td>99.0</td>
<td>99.9</td>
</tr>
<tr>
<td>UV\textsubscript{254}cm\textsuperscript{-1}</td>
<td>0.119</td>
<td>0.087</td>
<td>0.086</td>
<td>0.089</td>
<td>0.006</td>
</tr>
<tr>
<td>% removal</td>
<td>53.8</td>
<td>63.9</td>
<td>36.5</td>
<td>54.9</td>
<td>66.7</td>
</tr>
</tbody>
</table>

4.2. Flux reduction of feed solutions with different particle compositions

The clean-water flux was measured in the range 4000–4300 L/m\textsuperscript{2}/h. Directly after the feed solutions entered the batch cell microfiltration reactor, the flux decreased rapidly to 245–1170 L/m\textsuperscript{2}/h. As filtration continued, the flux fell to 55 L/m\textsuperscript{2}/h, as shown in Figure 2. The flux reduction was larger with raw sludge, as expected. Surprisingly, the flux reduction with the centrifuged solutions was similar to the raw sludge, although the solid concentrations of the solutions (38.2–63.9 NTU) were significantly lower than that of raw sludge (580 NTU). In addition, the flocs remaining at the bottom of the centrifuge tubes were re-suspended with distilled water and used in the microfiltration. As shown in Figure 2, the flux reduction for the flocs was relatively low, indicating that the effects of large particles on membrane fouling were less detrimental. The flocs solution had the largest solid concentration (19,067 mg/L). It has been reported that supernatant is the main contributor to membrane fouling in MBR reactors (Y.J. Liu et al., 2011). Supernatant caused 50% of specific cake resistance (Nuengjamnog et al., 2005) and was the main contributor to membrane fouling.
4.3. Flux recovery by enhanced-backwash cleaning

Enhanced backwashing uses chemicals during the backwash process. In this study, enhanced backwash was simulated with physical cleaning by stirring followed by chemical cleaning by NaOCl soaking (100 mg Cl₂/L). The flux recovery of each cleaning was investigated for solutions of raw sludge, supernatant from solutions centrifuged at various rotational speeds, and for the remaining flocs, and the results are shown in Figure 3. The physical cleaning was effective for the recovery of the lost flux by filtration of the AnMBR sludge. The flux recovery by physical cleaning was at most approximately 2225 L/m²/h. Chemical cleaning could increase the flux recovery. Chemical cleaning showed greater recovery with the feed solution treated at 1500 rpm. No recovery by chemical cleaning was observed for the solution treated at 500 rpm. This result is consistent with the result of the poorest flux reduction being for the solution treated at 500 rpm. The good recovery achieved by the cleaning methods using centrifuged supernatants implies that the fouling layer made by cake formation could be effectively removed by enhanced backwash. However, the lower recovery of the flocs solution showed that substantial concentrations of solids might disrupt chemical cleaning, and thus, removal of solids would be required for enhanced backwash to be applied.
**Figure 3.** Flux recovery by each cleaning method ($J_0$: clean water flux, $J_f$: flux at the end of each run, $J_{ph}$: flux recovered by physical cleaning, $J_{ch}$: flux recovered by NaOCl cleaning)

Resistances after each process were calculated based on the equation given in section 2 and are shown in Table 4. The intrinsic membrane resistances, which are considered as characteristics of the PTFE membrane, were similar in all cases. $R_T$ of the raw sludge had a higher value (73.94 × 10$^{10}$ m$^{-1}$) than those of the supernatants. The resistances recovered by physical cleaning, $R_{ph}$, accounted for the majority of $R_T$ (96~98%). The supernatants exhibited lower recoveries than raw sludge in surface fouling.

**Table 4.** Membrane resistances, resistances recovered by physical and by chemical cleaning, and resistance by irreversible fouling of PTFE filters ($R$ × 10$^{10}$, m$^{-1}$)

<table>
<thead>
<tr>
<th>Feed water</th>
<th>$R_m$</th>
<th>$R_{ph}$</th>
<th>$R_{ch}$</th>
<th>$R_{ir}$</th>
<th>$R_T$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Raw sludge</td>
<td>0.97</td>
<td>73.61</td>
<td>0*</td>
<td>0.47</td>
<td>73.94</td>
</tr>
<tr>
<td>SC500</td>
<td>0.99</td>
<td>56.47</td>
<td>0*</td>
<td>1.08</td>
<td>57.40</td>
</tr>
<tr>
<td>SC1000</td>
<td>0.96</td>
<td>49.29</td>
<td>0.35</td>
<td>0.55</td>
<td>50.18</td>
</tr>
<tr>
<td>SC1500</td>
<td>0.96</td>
<td>49.58</td>
<td>1.08</td>
<td>0.67</td>
<td>51.34</td>
</tr>
<tr>
<td>Flocs</td>
<td>0.90</td>
<td>21.40</td>
<td>4.38</td>
<td>18.54</td>
<td></td>
</tr>
</tbody>
</table>

*No effect of cleaning treatment observed

5. CONCLUSIONS

In this study, the effect of sludge properties in terms of particle composition on the performance of enhanced backwash was investigated. In addition, fouling mechanisms for specific feed solutions with different particle compositions were evaluated by flux recovery and resistances. Particle size distributions were classified by three centrifugal conditions: 500, 1000, and 1500 rpm.

The flux reductions for the three supernatants were significant. The flux with the solution treated at 500 rpm was lowest and two solutions treated at 1000 and 1500 rpm were similar, which implies that certain ranges of particle size are particularly important for fouling in AnMBRs. The maximum flux recovery was found to be approximately 2225 L/m$^2$/h by physical cleaning and 2743 L/m$^2$/h by NaOCl cleaning. The efficiency of enhanced backwash was lowest for the solution centrifuged at 500 rpm. Particle composition such as fine-particle concentration played an important role in fouling and the chemical cleaning efficiency. Further research into the cleaning efficiency of the proposed technology is required to determine the optimal values of duration and dose.
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