Membrane Fouling in a Membrane Bioreactor (MBR): Sludge Cake Formation and Fouling Characteristics

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Abstract: A submerged membrane bioreactor (MBR) with a working volume of 1.4 L and a hollow fiber microfiltration membrane was used to treat a contaminated raw water supply at a short hydraulic retention time (HRT) of ~1 h. Filtration flux tests were conducted regularly on the membrane to determine various fouling resistances, and confocal laser scanning microscopy (CLSM) and scanning electron microscopy (SEM) were employed to characterize the biofouling development and sludge cake formation on the membrane. The experimental results demonstrate that the MBR is highly effective in drinking water treatment for the removal of organic pollutants, ammonia, and UV absorbance. During the MBR operation, the fouling materials were not uniformly distributed on the entire surface of all of the membrane fibers. The membrane was covered partially by a static sludge cake that could not be removed by the shear force of aeration, and partially by a thin sludge film that was frequently washed away by aeration turbulence. The filtration resistance coefficients were $308.4 \times 10^{11}$ m$^{-1}$ on average for the sludge cake, $32.5 \times 10^{11}$ m$^{-1}$ on average for the dynamic sludge film, and increased from $10.5 \times 10^{11}$ to $59.7 \times 10^{11}$ m$^{-1}$ for the membrane pore fouling after 10 weeks of MBR operation at a filtration flux of $0.5 \text{m}^3/\text{m}^2\cdot\text{d}$. Polysaccharides and other biopolymers were found to accumulate on the membrane, and hence decreased membrane permeability. More important, the adsorption of biopolymers on the membrane modified its surface property and led to easier biomass attachment and tighter sludge cake deposition, which resulted in a progressive sludge cake growth and serious membrane fouling. The sludge cake coverage on the membrane can be minimized by the separation, with adequate space, of the membrane filters, to which sufficient aeration turbulence can then be applied. © 2005 Wiley Periodicals, Inc.

Keywords: confocal laser scanning microscopy (CLSM); extracellular polymeric substances (EPS); filtration resistance; membrane bioreactor (MBR); membrane fouling; transmembrane pressure (TMP)

INTRODUCTION

A membrane bioreactor (MBR), which consists of an activated sludge (AS) reactor and a microfiltration/ultrafiltration membrane, is a promising biological wastewater treatment technology. It takes advantage of rapid developments in membrane manufacturing and has the potential to fundamentally advance biological treatment processes. With its unique features of excellent effluent quality, ensured biomass–water separation, small footprint demand, better operational control of sludge concentration, and other biological conditions, MBR is increasingly being used in wastewater treatment and the reclamation of treated effluents (Stephenson et al., 2000; Thomas et al., 2000; Visvanathan et al., 2000). Recently, MBR has been demonstrated to be highly effective for the drinking water treatment of polluted surface water supplies (Li and Chu, 2003). Its potential for water and wastewater treatment lies not only in its application to biological degradation and nitrification, but also because it could replace other conventional treatment units, including flocculation, sedimentation, filtration, and disinfection.

Despite these advantages and potential, membrane fouling is still the major problem that hinders the practical application of MBRs. It has been found that membrane performance varies considerably with the properties of AS. The composition and characteristics of the MBR suspension affect biofouling development and sludge cake deposition on the membrane surface (Brindle and Stephenson, 1996; Defrance et al., 2000; Hong et al., 2002; Lee et al., 2003). Half of the total fouling resistance is due to soluble compounds, especially biopolymers, in the MBR sludge (Wisniewski and Grasmick, 1998; Huang et al., 2000). In addition to soluble microbial products (SMPs), extracellular polymeric substances (EPSs) also play an important role in sludge cake accumulation on the membrane surface (Brindle and Stephenson, 1996; Chang and Lee, 1998; Cho and Fane, 2002; Rosenberger and Kraume, 2002). However, previous investigations have
often overlooked the nonuniformity in the distribution of the fouling materials on the membrane surface in an MBR. Different fouling components, such as pore fouling, thin sludge layer fouling, and thick sludge cake fouling have not been well distinguished or characterized. Many reports focus on the fact that a high SMP and EPS content increases the viscosity of the AS mixed liquor, and hence reduces its filterability through a membrane (Nagaoka et al., 1996; Chang et al., 2001). The more important effects of biopolymers in the AS suspension on sludge cake attachment to the membrane surface remains to be characterized.

The aims of the present experimental study with a submerged MBR were to develop a new fouling model to address the feature of partial sludge cake coverage on the membrane surface, to quantify the resistance coefficients of different fouling forms, including pore fouling, dynamic sludge film, and static sludge cake resistances, and to characterize the interaction between biopolymers and bacteria that attach to the membrane surface, and the effect of this interaction on sludge cake formation using confocal laser scanning microscopy (CLSM) and scanning electron microscopy (SEM).

MATERIALS AND METHODS

MBR Set-Up

A submerged membrane reactor was used for the biofouling study. The set-up and operation of the MBR was similar to that previously described (Li and Chu, 2003) except that the reactor size was different. In brief, two polyethylene (PE) hollow-fiber membrane modules (pore size = 0.4 μm, surface area = 0.03 m², Mitsubishi Rayon) were immersed inside an activated sludge reactor to form an MBR. The reactor was made of acrylic plate with dimensions 15 × 8 × 20 cm³ (L × W × H) and a working volume of 1.4 L at a water depth of 12 cm. A level controller was used to operate the feeding pump and the effluent was drawn from the MBR by a suction pump. Aeration was provided through a porous air diffuser at the bottom to generate strong turbulence to continuously clean the surface of the membrane. A manometer was mounted between the membrane modules and the suction pump to monitor the transmembrane pressure (TMP).

The influent to the MBR was a simulated water supply that was contaminated by sewage discharge. Domestic sewage collected from a full-scale wastewater treatment plant was added to tap water at a ratio of around 1:10 to simulate water pollution (Li and Chu, 2003). The water sample was stabilized for 2–3 days under ambient conditions before being fed into the MBR. The influent had a total organic carbon (TOC) content of ~4 mg/L, and NH₄Cl was frequently added to increase the NH₃-N concentration to ~3 mg/L. With a highly diluted influent, a short hydraulic retention time (HRT) of around 1 h was approached and maintained. During stable operation, the membrane modules were cleaned approximately once a week by flushing briefly for 30 sec with running tap water. A chemical backwash using NaOCl (1.5% free chlorine) was conducted once during the study period. The general parameters of the MBR in its stationary operation are summarized in Table I.

Filtration Resistance Determination

The filtration flux through a uniform membrane surface in an MBR can be described by the general form of Darcy’s law (Duranceau, 2001),

\[ J = \frac{\Delta P}{\mu(R_m + R_f + R_s)} \]

where \( J \) is the permeation flux, \( \Delta P \) is the TMP that is applied, \( \mu \) is the viscosity of the permeat, and \( R \) is the total resistance, which is the combination of the three resistances of \( R_m \), the intrinsic membrane resistance, \( R_f \) the pore fouling resistance caused by solute adsorption on the wall of the pores, and \( R_s \), the resistance of the sludge layer deposited on the membrane surface.

Due to the continuous cleaning action of the turbulence shear caused by aeration in an MBR, the membrane surface may not be fully covered by a stable sludge cake. A new parameter, sludge cake coverage, \( \eta_c \), is defined here as the ratio of the membrane surface area that is covered by a static sludge cake to the total membrane surface area, or \( \eta_c = A_{cake}/A_{total} \) (Fig. 1). For the remaining membrane surface area that is not permanently covered by a static sludge cake, the filtration of the sludge suspension will result in continuous biomass deposition. The biomass forms a thin sludge film that temporarily attaches to the membrane surface and can be promptly removed by aeration. The filtration resistance of this thin sludge film, \( R_{sf} \), is less than that of the stable sludge cake, \( R_{sc} \). Thus, the overall MBR permeation flux through a membrane that is covered partially by a dynamic sludge film and partially by a static sludge cake can be written as \( J = J_{sf} + J_{sc} \), where \( J_{sf} \) is the flux through the thin sludge film and \( J_{sc} \) is the

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>MLSS (mg/L)</td>
<td>2,500–3,000</td>
</tr>
<tr>
<td>MLVSS (mg/L)</td>
<td>2,000–2,500</td>
</tr>
<tr>
<td>HRT (hr)</td>
<td>0.9–1.2</td>
</tr>
<tr>
<td>SRT (day)</td>
<td>48–70</td>
</tr>
<tr>
<td>Flux (m³/m²·d)</td>
<td>0.45–0.55</td>
</tr>
<tr>
<td>Organic loading (g TOC/g VSS·d)</td>
<td>0.089–0.112</td>
</tr>
<tr>
<td>NH₃-N loading (g/g VSS·d)</td>
<td>0.057–0.098</td>
</tr>
<tr>
<td>Dissolved oxygen (mg/L)</td>
<td>6.1–8.1</td>
</tr>
<tr>
<td>pH</td>
<td>6.1–7.8</td>
</tr>
<tr>
<td>Temp (°C)</td>
<td>15–25</td>
</tr>
<tr>
<td>Suction pump on/off (min)</td>
<td>18/2</td>
</tr>
</tbody>
</table>
flux through the sludge cake. Hence, Eq. 1 can be further modified to:

\[ J = \frac{\Delta P(1 - \eta_c)}{\mu(R_m + R_f + R_{sf})} + \frac{\Delta P\eta_c}{\mu(R_m + R_f + R_{sc})}. \]  

(2)

For a new membrane in deionized (DI) water with a viscosity of \( \mu_w \), \( \eta_c = 0 \), \( R_f = 0 \), and \( R_{sf} = 0 \); Eq. 2 becomes:

\[ \Delta P = \mu_w R_m J. \]  

(3)

For a used membrane in DI water without sludge cake coverage after physical cleaning, \( \eta_c = 0 \) and \( R_{sf} = 0 \), Eq. 2 becomes:

\[ \Delta P = \mu_w (R_m + R_f) J. \]  

(4)

For a used membrane without sludge cake coverage after physical cleaning in the MBR sludge suspension, \( \eta_c = 0 \) at the initial filtration stage, the permeate effluent has a viscosity of \( \mu_e \), Eq. 2 becomes:

\[ \Delta P = \mu_e (R_m + R_f + R_{sf}) J. \]  

(5)

For a membrane fully covered by sludge cake and submerged in DI water, \( \eta_c = 1 \), Eq. 2 becomes:

\[ \Delta P = \mu_w (R_m + R_f + R_{sc}) J. \]  

(6)

Therefore, the different filtration resistances, \( R_m \), \( R_f \), \( R_{sf} \), and \( R_{sc} \), can be estimated through filtration flux tests by the measurement of the change in the TMP against an increasing flux under different fouling and filtration conditions. Before the regular physical cleaning of the membrane, aeration in the MBR was reduced to a minimum level overnight to allow a full coverage of sludge cake to develop on the membrane surface. The filtration test was then conducted for the fouled membrane in DI water, which corresponded to the scenario of Eq. 6. After a physical wash removing the sludge cake, flux tests were conducted for the clean membrane, first in DI water, which fitted with the filtration condition of Eq. 4, and then immediately in the MBR with aeration provided, which approximated the situation of Eq. 5.

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**Microscopic Observation of MBR Membrane Fouling**

Membrane fibers were cut from the modules in the MBR and examined microscopically by confocal laser scanning microscopy (CLSM) (LSM Pascal, Zeiss, Thornwood, NY) and scanning electron microscopy (SEM) (360, Cambridge Stereoscan, Cambridge UK). The bacterial population and associated biopolymers attached to the membrane were observed using the CLSM equipped with three lenses, i.e., Fluar 10X/NA0.5, Plan-Neofluar 40X/NA0.75, and a Plan-Apochromat 63X/NA1.4 (Pawley, 1995; Zhang and Fang, 2001). For bacterial and biopolymer staining, two probes were collectively applied: SYTO9 (25 \( \mu \)M, Molecular Probes, Eugene, OR) to target all bacteria, and ConA-TRITC (250 mg/L, Molecular Probes) to target the polysaccharides with D-glucose or D-mannose. The membrane specimens were placed and stained in slide wells that were incubated in a moisture chamber (a 50-ml conical centrifuge tube, Corning, Corning, NY) at room temperature for 30 min. After staining with ConA, all of the slide wells were washed gently three times with a phosphate buffer saline solution to remove any unbound probes. Detection of the probes was performed using an optical filter set that consisted of an HFT 488 nm/545 beam splitter with a 488 nm laser beam, a NFT545 beam splitter, and a BP505-530 bandpass filter for the bacteria stained by SYTO9, in combination with the setting of 543 nm/NFT545/LP560 for the polysaccharides stained by ConA-TRITC.

For SEM observation, the membrane samples were prepared following the method described by Diao et al. (2004). In brief, a membrane fiber was first washed with a 0.1 M phosphate buffer solution at pH = 7.2, then placed in the buffer solution with 2.5% glutaraldehyde for 10 h. The sample was dehydrated by immersion in a series of ethanol solutions of 50%, 75%, 85%, 95%, and 100%, for 15 min at each ethanol concentration. The dried membrane fiber was fixed on a filter paper and sputter-coated with gold-palladium before the membrane surface and its sludge attachment were examined under the SEM.

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**Analytical Methods**

The organic content of the water was quantified by a TOC analyzer (TOC-5000A, Shimadzu, Kyoto, Japan) using the combustion-infrared method. The viscosity of the liquid was measured with a viscosity meter (Visco Star-L, Fungilab). Ammonia nitrogen was analyzed by the electrical-chemical method using an ammonia-selective electrode (95-12, Orion, Cambridge, MA) with an electrometer (920A, Orion). The UV absorbance of the water sample at 254 nm, \( \text{UV}_{254} \), was determined by a UV/VIS spectrometer (Lambda 12, Perkin Elmer, Norwalk, CT). The mixed liquor suspended solid (MLSS) and mixed liquor volatile suspended solid (MLVSS) of the sludge suspension in the MBR were measured regularly according to the Standard Methods (APHA, 1998).

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**Figure 1.** Schematic diagram of partial sludge cake coverage on the membrane surface in an MBR.
RESULTS

Water Treatment Efficiencies

The submerged MBR for the treatment of a contaminated raw water supply was continuously operated for more than 150 days. The treatment performance was nearly identical to that previously reported for a larger MBR serving a similar purpose (Li and Chu, 2003). During its stationary operation at a short HRT of about 1 h, the MBR removed nearly 60% of the organic impurities from the water and decreased the TOC from 4.35 ± 1.27 to 1.78 ± 0.42 mg/L. UV$_{254}$ was reduced by more than 60% from ~9.8 ± 1.9 to 3.6 ± 0.7 m$^{-1}$, and more than 95% of the NH$_3$-N was removed by biological nitrification (Fig. 2). Although the influent had a rather low pollutant strength, a short HRT of 1 h effectively increased the substrate loading to the MBR, producing a sufficient biomass in the reactor (Table 1). The experimental results suggest that the MBR process is feasible for use in drinking water treatment.

Membrane Filtration Resistances

With the elevated effluent flux used at a short HRT, the membrane fouling problem of the MBR was well demonstrated by the increase in TMP (Fig. 3). When the TMP reached a level of around 50 kPa, the membrane modules had to be cleaned physically by flushing with tap water to remove the sludge cake on the membrane surface. An MBR operation that usually lasted for a week between two physical cleaning procedures was termed an operational stage. At the end of stage 10, the membranes were washed chemically with NaClO to show the effect of chemical cleaning. During a working stage with an effluent flux of around 0.5 m$^3$/m$^2$/d, the TMP increased at an average rate of 4.0–4.5 kPa/d. The initial TMP after a physical flushing, TMP$_\text{p}$, increased 1.0–1.5 kPa with each running stage, indicating a decrease in the membrane permeability. After a chemical wash, the TMP$_\text{p}$ dropped dramatically and the filtration capacity of the membrane was almost fully recovered.

Filtration flux tests were conducted regularly for the membrane in DI water ($\mu_\text{w} = 0.96 \times 10^{-3}$ Pa·s) and the AS suspension ($\mu_\text{e} \approx 1.03 \times 10^{-3}$ Pa·s) to determine the individual resistance coefficients defined above. The TMP-flux test results for the membrane in stage 7 are presented in Figure 4. The slopes of the TMPs vs. the fluxes give the resistance coefficients in relation to the filtration cases of Eqs. 3–6. A new membrane had a resistance of $R_\text{m} = 1.5 \times 10^{11}$ m$^{-1}$. At the end of stage 7, the membrane had a fouling resistance of $R_\text{f} = 37.4 \times 10^{11}$ m$^{-1}$. The resistance of the sludge cake that formed on the membrane was calculated as $R_\text{sc} = 305.7 \times 10^{11}$ m$^{-1}$, which was much higher than the fouling resistance. A slightly curved TMP increase with the flux was observed for the membrane that was placed in the MBR suspension after a physical wash. This was probably caused by the progressive formation of the sludge film on the membrane surface. From the upper portion of the data, the resistance of the thin sludge film was estimated as $R_\text{sf} = 29.6 \times 10^{11}$ m$^{-1}$.
With the individual resistances that were determined by the filtration tests, the sludge cake coverage, \( \eta_c \), by the end of each running stage was estimated from Eq. 2 (Fig. 5). Throughout the MBR operation, it was found that \( R_{sf} \) and \( R_{sc} \) did not change considerably with the running time (Fig. 5) from the respective average values of \( 32.5 \times 10^{11} \) and \( 308.4 \times 10^{11} \) m\(^{-1} \). The filtration resistance due to pore fouling increased significantly after 10 stages from \( 10.5 \times 10^{11} \) to \( 59.7 \times 10^{11} \) m\(^{-1} \), and dropped to \( 16.4 \times 10^{11} \) m\(^{-1} \) after the chemical wash. The sludge cake coverage on the membrane before each physical cleaning ranged from 51–57%. The physical wash was able to effectively remove the sludge cake attached to the membrane surface, and thus the filtration capacity was largely recovered.

Microscopic Observation of Membrane Fibers

Partial sludge cake coverage on the membrane surface in a working MBR is well demonstrated by microscopic examination (Figs. 6, 7). The CLSM image of a typical membrane fiber showed a sludge cake of around 100 \( \mu \)m in thickness on the top surface of the fiber, but no sludge deposition on the bottom of the fiber (Fig. 6a). After a physical wash with running tap water, the sludge cake was removed and a smooth membrane surface was recovered (Fig. 6b). Using different CLSM probes, both bacteria (green) and polysaccharides-based organic polymers (red) were identified. Bacteria aggregated into clusters to form the sludge cake on the membrane surface and polysaccharides were found between the bacterium clusters (Fig. 6c). The interaction between the biomass and biopolymers within the sludge cake was similar to that previously reported for the suspended flocs of activated sludge (Liao et al., 2001; Li et al., 2003). After a physical wash to remove the sludge cake, polysaccharides were still left on the membrane surface, together with a few bacteria (Fig. 6d). Inside the membrane fiber, 100 \( \mu \)m below the surface scanned by the CLSM, polysaccharides were also detected (Fig. 6e). The CLSM photos indicate that biopolymers can be adsorbed onto the membrane surface and intrude into the membrane pores, which cannot be readily removed by regular physical washing. The accumulation of polysaccharides and other biopolymers increased the fouling resistance, particularly \( R_f \), as shown above by the filtration tests (Fig. 5). More important, the biopolymer residue on the membrane would modify the membrane surface property and make it friendlier to bacterium attachment, leading to easier biomass accumulation and sludge cake formation during the MBR operation. Therefore, chemical cleaning has to be applied periodically to remove biopolymers and other colloid residues from the membrane.

SEM examination also revealed the biofouling problem and the partial sludge cake coverage on the membrane surface (Fig. 7a). Bacteria were apparently bridged by biopolymers to form the sludge cake attached to the membrane (Fig. 7b). After a physical wash, the sludge cake was removed, but there was still a slime layer deposited on the membrane (Fig. 7c). This slime layer appeared to grow in thickness with the MBR operation, and eventually the membrane surface was almost fully covered by the gel-like slime material (Fig. 7d). After a chemical wash with NaClO, the gel-like slime layer was completely removed (Fig. 7e), which resulted in a much lower fouling resistance, as indicated above by the filtration tests (Fig. 5).

DISCUSSION

Sludge Cake Formation and Removal

As \( R_m \) is much less than the other resistances, and \( R_{sf} \approx R_{sc} \), Eq. 2 can be simplified to:

\[
J \approx \frac{\Delta P}{\mu_e (R_f + R_{sf})} \left( 1 - \frac{R_{sc}}{R_{sc} + R_f} \eta_c \right).
\]

\( \Delta P \) is the transmembrane pressure, \( \mu_e \) is the effective viscosity of the sludge cake, and \( J \) is the filtration flux.
When the pore fouling is not serious, or $R_f \ll R_{sc}$, Eq. 7 can be approximated to:

$$\Delta P = \frac{\mu(R_f + R_{sf})Q}{(1 - \eta_c)A},$$

where $A$ is the total membrane surface area and $Q$ is the water production rate of the MBR. Hence, an accumulation of a sludge cake, i.e., an increase of $\eta_c$, during the MBR operation will result in a continuous increase in the TMP that is required to maintain a certain effluent production rate. Therefore, the minimization of the cake coverage by aeration turbulence is essential to the MBR operation and water production.

Two opposite actions regulate the rate of sludge cake formation on the membrane surface: sludge attachment, which is driven by the suction filtration, and sludge removal, which is caused by the fluid shear of aeration turbulence. With an apparent filtration flux of $J$, the sludge deposition rate on the membrane surface is $JC$, where $C$ is the sludge concentration. However, if the membrane is partially covered by a sludge cake that is nearly impermeable to the effluent, then the rate of sludge deposition on the uncovered membrane surface area can be written as:

$$S_d = \frac{JC}{1 - \eta_c}.$$  \hspace{1cm} (9)

The sludge removal process is more complex. The force of sludge removal by air-scouring turbulence is enhanced by the aeration flowrate, $F$. The difficulty of sludge detachment, however, depends on the stickiness of the biomass, mainly bacteria, $\alpha$, which has a maximum value of 1. It can be anticipated that the potential amount of sludge that
can be removed from the membrane will decrease as the bacterial stickiness increases. In simplified terms, the maximum potential rate of sludge removal from the membrane by aeration may be described by:

\[ S_r = c(1 - \alpha)F^b, \]  

(10)

where \( b \) and \( c \) are constants for a given MBR system. An increase in the stickiness of the biomass, which is largely affected by the bacterial surface properties, will worsen the problem of sludge cake formation on the membrane. However, a higher intensity in shear turbulence by aeration lessens the cake fouling problem to a considerable extent (Bian et al., 2000; Le Clech et al., 2003; Yu et al., 2003).

If \( S_r > S_d \) for the entire membrane surface, then there will be no permanent sludge cake deposition on any part of the membrane, although temporal biomass attachment always occurs during the MBR suction filtration. In actuality, however, the shear force of aeration turbulence is not uniformly exerted on the surface of all of the membrane filters placed in the MBR. Some areas of the membrane, such as those within the string of membrane fibers, may not experience strong turbulence, even at an elevated aeration rate. For such areas where \( S_r < S_d \), sludge attaches to the surface and builds up to form a stable sludge cake. This sludge cake formation reduces the membrane surface area available for effluent filtration. Consequently, as suggested by Eq. 9, the sludge deposition rate on the area without cake coverage increases, and the force of the sludge removal is unchanged. This would result in a progressive growth of sludge cake coverage, leading to serious membrane fouling. Thus, sludge cake fouling on the membrane

Figure 7. SEM photographs of membrane fouling and sludge cake coverage: (a) a membrane fiber with partial sludge cake coverage before a physical wash; (b) sludge cake deposited on the membrane; (c) membrane (100 days old) surface after a physical wash; (d) membrane (500 days old) surface after a physical wash; and (e) membrane surface after a chemical wash using NaClO.
surface is hardly a reversal process unless physical cleaning is exercised. Nonetheless, for an MBR with a low filtration flux, $S_d < S_r$, can be maintained for a large portion of the membrane, even though a part of the membrane has been covered by the sludge cake. A stable operation status can be established for MBR filtration without the continuous growth of sludge cake coverage.

**Effect of Biopolymer Adsorption on Sludge Cake Formation**

The influence of biopolymers on MBR fouling has been a subject of investigations that focus mainly on the role of extracellular polymeric substances (EPSs) (Nagaoka et al., 1996; Chang and Lee, 1998; Bouhabila et al., 2001; Rosenberger and Kraume, 2002). The specific resistance of EPS, which is defined as the filtration resistance ($m^{-1}$) divided by the EPS density on the membrane (kg-TOC/m²), is on the order of $10^{16}$ to $10^{17}$ m/kg (Nagaoka et al., 1996).

The composition of the mixed liquor in an MBR, specially the EPS abundance, also affects the filterability of the sludge suspension. The higher the suspended EPS concentration, the lower the filtration flux. The filtration index $I_{40}$, which is the sludge permeate flux after 40 min compared with the clear water flux, was found to decrease by 80% when the suspended EPS increased from 15 to 90 mg/L (Rosenberger and Kraume, 2002). It is understood that an increase of biopolymer materials in the sludge suspension increases its viscosity, and hence reduces the MBR permeation flux (Nagaoka et al., 1996; Chang et al., 2001). The present findings, however, place more emphasis on the adsorption of biopolymers on the membrane and its effect on sludge cake formation. The deposition of polysaccharides and other biopolymers increases the stickiness of the membrane surface, which allows easier and faster bacterial adhesion. In addition, the EPS holds the biomass more tightly on the membrane and increases the difficulty of sludge removal by aeration turbulence.

It should be noted that the CLSM procedure was employed only for visualization of bacterial cells and associated substances on the membrane fibers, rather than for identification of any particular materials. Although ConA-TRITC, a fluorescently labeled lectin, was used to target polysaccharides, the red-colored region in the CLSM images did not exclude the presence of other biopolymers and organic molecules (Fig. 6). It has been found that lectins bind not only to polysaccharides that contain their inhibition sugars, such as D-glucose, but also to other organics, such as bacterial alginate. Alginate can be involved in the interaction between ConA and polysaccharides, although this type of molecule apparently contains none of the ConA inhibition sugars (Strathmann et al., 2002). However, such possible interferences to the fluorescent staining do not undermine the findings of the present CLSM visualization for the fouling materials and sludge cake formation on the MBR membrane. The red color in the CLSM photos is a general representative of biological products, e.g., EPS and SMP, that were associated with bacterial cells or attached to the membrane fibers. The occurrence of polysaccharides also suggests the presence of proteins. Proteins, which were not targeted in the present microscopic visualization, are another important EPS component (Liu and Fang, 2003). They are macromolecules with both hydrophobic and hydrophilic properties. They could attach to the membrane surface more tightly, leading to the fouling problem. Although proteins did not appear to be the predominant EPS constituent in the MBR sludge compared to polysaccharides (data not shown), the exact role of proteins in the attachment of fouling matter and sludge cake growth on the membrane remains to be identified.

Based on the filtration resistance estimation and microscopic observation, the following MBR fouling mechanism can be summarized. Biopolymers in the MBR sludge suspension, especially polysaccharides, stick to the membrane surface and intrude into the membrane pores. The accumulation of biopolymers on the membrane, which cannot be readily removed by aeration turbulence or regular physical flushing, gradually increases the fouling resistance. More important, biopolymer adsorption on the membrane surface leads to the easier attachment of bacteria and the formation of a sludge cake on the membrane where the shear force of aeration cleaning is relatively weak. The partial sludge cake coverage on the membrane will increase both the filtration flux and the sludge deposition rate on the uncovered membrane surface area. This leads to a progressive growth of sludge cake on the membrane, causing a considerable increase in TMP and a decrease in water production.

Aeration turbulence is an effective means of removing the sludge cake from the membrane surface during the MBR operation. A high aeration rate certainly can reduce sludge attachment on the membrane, but it also increases the operation cost. A tall MBR is preferred to increase the aeration intensity without increasing the air flowrate. To improve the effectiveness of membrane cleaning by aeration, membrane fibers should be well separated and spaced without close contact. Hence, the entire surface area of all membrane fibers can be cleaned constantly by the turbulent shear of aeration to minimize the formation of a sludge cake.

**CONCLUSIONS**

MBR is highly effective in the treatment of polluted water supplies to remove organic pollutants, ammonia, and UV absorbance. However, for a highly diluted influent, an increase in the MBR filtration flux at a short HRT could considerably increase the TMP and accelerate the fouling problem. It was found that the fouling materials did not distribute uniformly on the surface of all of the membrane fibers. This was probably caused by the reduction in the shear force of aeration turbulence to a certain portion of the membrane surface in the MBR. The membrane was covered partially by a static sludge cake and partially by
a thin sludge film. The filtration resistance coefficients were $308.4 \times 10^{11} \text{ m}^{-1}$ on average for the sludge cake, $32.5 \times 10^{11} \text{ m}^{-1}$ on average for the dynamic sludge film, and increased from $10.5 \times 10^{11}$ to $59.7 \times 10^{11} \text{ m}^{-1}$ for the pore fouling after 10 weeks of MBR operation. According to CLSM and SEM observations, polysaccharides and other biopolymers accumulated on the membrane, and hence decreased the membrane permeability. Biopolymers in the MBR suspension also increased the liquid viscosity and reduced its filterability. More important, the adsorption of biopolymers on the membrane modified its surface property and led to easier biomass attachment and tighter sludge cake deposition, which resulted in a progressive growth of sludge cake on the membrane surface. Sludge cake coverage on the membrane could be minimized by separating the membrane filters with adequate space between them, to allow sufficient shear of aeration turbulence for cleaning.

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References


