Permeate Characteristics and Operation Conditions of a Membrane Bioreactor with Dispersing Modified Hollow Fibers

Choon Hwan Shin† and Robert Johnson

Department of Environmental Engineering, Dongseo University, Pusan, Korea
School of Physical & Chemical Science, Queensland University of Tech., 2 George St. Brisbane Australia 4001

Received May 24, 2006; Accepted September 27, 2006

Abstract: In this study, the effects of the pretreatment method and the dispersion of hollow fiber membranes on the permeate flux were investigated. As the ethanol (as solvent) concentration increased from 10 to 90 %, the permeation rate through the polysulfone membrane increased. The permeate flux was constant at concentrations greater than 20 %. A relatively high flux was obtained at moderately low pressures and the pressure increase rate decreased with the dispersion of the membrane. As a packing density decreased, the permeate flux increased, resulting in low pressure. The maximum permeate flux (30.08 threads/cm²) was achieved under stable conditions after 4 days’ separation. On the other hand, the operation conditions were determined in a submerged membrane bio-reactor using dispersed hollow fiber membranes. From permeate flux measurements according to the packing density, we found that 20 membrane modules showed a stable and high flux. The growth and decay coefficients of microorganisms were 0.6464 and 0.0712, respectively. From the data analysis, we determined that the most efficient conditions for long-term operation were an MLSS concentration within the range from 3800 to 8200 mg/L and organic compound loadings from 300 to 600 mg/L.

Keywords: permeate flux, MBR, hollow fiber, operation condition, growth and decay coefficient

Introduction

Because of improvements in membrane separation techniques, a recent series of studies was conducted on water/wastewater treatment systems in conjunction with membrane separation [1-3]. One of the examples is a combination of biological secondary treatment with an MF process. Because this process does not produce any bacilli, such as a colon bacillus, as well as SS, resulting in a BOD 5 of 1~5 mg/L, its effluent can be recycled as indoor gray water for cleaning and toilet reuse. Moreover, its energy consumption is 0.3~0.4 kWh/m³, which is almost equivalent to that for secondary municipal wastewater treatment. MF processes with powdered BAC also have been developed to remove non-degradable soluble organics and bacteria hazardous to human beings. The combined processes, however, have intrinsic problems that must be resolved, such as membrane fouling and washing, recovery of permeate flux, and the cost of the membrane module itself. Therefore, much research and development effort has to be exerted before they come into practical use.

Among the applications of membrane separation in wastewater treatment, hollow fiber membranes (HFM) have been used for solid/liquid separation by directly submersing them in existing activated sludge tanks [4,5]. Because this process can maintain a high concentration of microorganisms in the activated sludge tank, it is suitable for municipal wastewater treatment with a high level of organics, even at low pressure (less than 0.5 kgf/cm²). Air bubbling in the system makes the hollow fiber membranes move around in the tank, providing a surface shear so that less membrane fouling, such as surface deposition and pore clogging, can be achieved. However, in any membrane separation system, it is inevitable to experience a lowering of the system ability due to membrane fouling, measurements of which have been a challenging task to resolve [6]. Membrane fouling is the irreversible deposition of solutes onto a membrane surface, caused by various factors, such as the feed water quality, the membrane

† To whom all correspondence should be addressed.
(e-mail: 6116shin@gdsu.dongseo.ac.kr)
Table 1. Specifications of the Hollow-Fiber Membrane Used in this Study

<table>
<thead>
<tr>
<th>Content</th>
<th>Characteristic</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pore size</td>
<td>0.01 ~ 0.02 µm</td>
</tr>
<tr>
<td>Type</td>
<td>Hollow fiber</td>
</tr>
<tr>
<td>Material</td>
<td>Polysulfone</td>
</tr>
<tr>
<td>Outer diameter of fiber</td>
<td>0.7 mm</td>
</tr>
<tr>
<td>Inner diameter of fiber</td>
<td>-</td>
</tr>
<tr>
<td>Membrane area</td>
<td>0.207 m²</td>
</tr>
</tbody>
</table>

Table 2. Operation Conditions of MBR

<table>
<thead>
<tr>
<th>Content</th>
<th>Condition</th>
</tr>
</thead>
<tbody>
<tr>
<td>Temp.</td>
<td>20 ± 1 °C</td>
</tr>
<tr>
<td>pH</td>
<td>6 ~ 7</td>
</tr>
<tr>
<td>Volume</td>
<td>14 L</td>
</tr>
<tr>
<td>DO</td>
<td>6 ~ 7 mg/L</td>
</tr>
<tr>
<td>MLSS</td>
<td>ca. 6000 mg/L</td>
</tr>
<tr>
<td>Air flow</td>
<td>15 L/min</td>
</tr>
</tbody>
</table>

Table 3. Composition of Synthetic Wastewater

<table>
<thead>
<tr>
<th>Chemical</th>
<th>Concentration (mg/L)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glucose (C₆H₁₂O₆)</td>
<td>1.000</td>
</tr>
<tr>
<td>KH₂PO₄</td>
<td>0.533</td>
</tr>
<tr>
<td>K₂HPO₄</td>
<td>0.640</td>
</tr>
<tr>
<td>(NH₄)₂SO₄</td>
<td>0.284</td>
</tr>
<tr>
<td>NH₄Cl</td>
<td>0.427</td>
</tr>
<tr>
<td>CaCl₂ 2H₂O</td>
<td>0.013</td>
</tr>
<tr>
<td>MgSO₄ 7H₂O</td>
<td>0.178</td>
</tr>
<tr>
<td>MnSO₄ H₂O</td>
<td>0.018</td>
</tr>
<tr>
<td>KCl</td>
<td>0.025</td>
</tr>
<tr>
<td>FeSO₄ 7H₂O</td>
<td>0.008</td>
</tr>
</tbody>
</table>

In this study, the optimum pretreatment conditions for the application of hollow fiber membranes were obtained with respect to the mixture solvent and pretreatment time. Next, to reduce membrane fouling, the permeate flow of the treated water and the transmembrane pressure (TMP) were investigated in terms of the dispersed placement of HFM modules. The permeation characteristics according to the packing density of HFM and the change of MLSS in aeration tanks were then studied to gain information regarding the optimum design of the HFM modules. Finally, in the application of HFM modules to solid/liquid separation of municipal wastewater, the operating conditions of the membrane bioreactor were determined to reduce membrane fouling by keeping the TMP as low as possible.

Methods

Module Manufacture and Operating Conditions

The hollow-fiber membranes (HFM) used in the submerged membrane module were prepared from polysulfone samples of 0.1 m or 0.01 ~ 0.02 m pore size, whose specifications are listed in Table 1. The PVC tubing and the HFM were 30 cm long. Polyurethane was used as a hardener.

The manufacturing procedure of the HFM module is shown in Figure 1. For convenience, it is described as 2 bundles, 4 bundles, and 10 bundles.

For the membrane dispersion experiments, the membranes were manufactured with various packing densities of the HFM in PVC tubing having a cross-sectional area of 1.33 cm². The membrane for each bundle had a 0.1 m pore size. The manufactured bundles were treated with 50% C₂H₅OH for ca. 1 h to make the membranes hydrophilic. Subsequently, the bundles were finally connected with a suction pump of 0.5 kgf/cm² in capacity. To study the filtration characteristics according to the membrane packing density and suction pressure, the permeate flux and the transmembrane pressure (TMP) were measured.
Figure 2. Schematic illustration of the experimental MBR.

Figure 3. Variations of the UV-spectral peak are after pretreatment.

The operating conditions of the submerged membrane bioreactor in this study are summarized in Table 2. The influent flow rate was controlled according to each membrane module to maintain a constant effective volume of 14 L. The membranes were operated to minimize fouling by repeating intermediate suction and stopping the suction pump with an attached timer. To the feed for microorganisms cultivation, synthetic wastewater was made in the laboratory and listed in Table 3.

The basic structure of the submerged MBR is summarized here. The manufactured HFM modules were installed in the middle of activated sludge tanks and connected to a suction pump (0.5 kgf/cm²). Air was provided through aeration tubes at the bottom of activated sludge tanks. A schematic diagram of the experimental set-up is shown in Figure 2.

Model Equations for Optimum MLSS Concentration of Submerged MBR

Model equations were used in this study to determine the growth coefficient (Y) and the decay coefficient (k_d) from the relationship between the biomass and the substrate concentration, which is given as

\[ \frac{1}{X_{AVG}} \cdot \frac{\Delta X}{\Delta t} = \frac{Y}{X_{AVG}} \cdot \frac{\Delta S}{\Delta t} - k_d \]  

where \( X \) = change of biomass concentration, mg/L
\( S \) = change of substrate concentration, mg/L
\( t \) = time increment (for one day, \( t = 1 \), day
\( X_{AVG} \) = average biomass concentration for \( t \), mg/L

With \( t = 1 \), the above equation is linearized as

\[ \frac{\Delta X}{X_{AVG}} = Y \cdot \frac{\Delta S}{X_{AVG}} - k_d \]  

By employing the yield coefficient (Y) and the endogenous decay coefficient (k_d) from Equation (2), and the results from Yamamoto et al. on organic wastewater treatment by using batch membrane reactors, the relationship between the optimum MLVSS, sludge retention time (SRT), and substrate loading can be obtained from the following equation:

\[ X = \frac{SRT}{HRT} \cdot \frac{Y(COD_i - COD_e)}{1 + k_d \cdot SRT} \]  

where \( X \) is the biomass concentration in the reactor and COD_i and COD_e are the COD concentrations of influent and effluent, respectively. Rearranging Equation (3) for substrate loading and SRT yields

\[ \frac{COD_i - COD_e}{X \cdot HRT} = \frac{1}{Y} \cdot \frac{1}{SRT} + \frac{k_d}{Y} \]  

With equation (4), the optimum MLSS concentration for continuous operation of the submerged MBR is derived.
Results and Discussion

Pretreatment Characteristics of Hollow-Fiber Membranes

For the case of an organic membrane, the performance and characteristics are significantly affected by the pretreatment methods. A 10∼90 % solvent mixture for pretreatment was used to obtain membrane permeate characteristics and effluent concentrations with respect to time. In the pretreatment processes, the extract was scanned in the range 200∼700 nm using a UV spectrometer. The scanning results indicated that as the solvent concentration increases, more extract can be produced near 200 nm and the extraction rate accelerates. At a solvent concentration over 70 %, continuous extraction was observed, even after a long period of operation. This situation may be due to membrane surface damage when the concentrations of remaining monomers (not involved in polymerization during membrane manufacture) and ethanol are too high [7, 8]. Figures 3 and 4 show the filtration results with various ethanol concentration (0∼70 %) and pretreatment methods. A certain degree of filtration ability was obtained with a solvent concentration above 20 %. However, for the case of a 70 % concentration, although the permeate flux increases with time, the SS removal efficiency is expected to decrease [7].

The change in the membrane surface was identified using an electron microscope. When the HFMs were submerged in the solvent mixture, the transformation of the membrane surface is illustrated in Figure 5, where SEM images are shown for the cases of 10, 20, and 60 % ethanol mixtures.

The surface pore size seems to increase with an increase in the amount of ethanol. However, according to Kamo, the SS removal rate is expected to decrease significantly. Thus, further study is required to satisfy this issue [7, 8].

The surface pore size decreases with 10 % ethanol and many small pores were observed with 20 % ethanol; with 60 % ethanol, however, there is a clear distribution of large and small pores. Therefore, as Kamo reported, the surface pore sizes of HFMs pretreated by solvent are controlled by deformation of the microfibril; this phenomenon is due to the surface tension of the water/ethanol mixture, whose average (m) can be expressed as follows:

\[
\sigma_m = \frac{\sigma_1 + \sigma_2}{\sigma_1 X_2 + \sigma_2 X_1}
\]

(5)

where \(\sigma\) and \(X\) are the surface tensions and the mole fraction, respectively. The subscripts 1 and 2 denote water and ethanol, respectively. Thus, the surface tension of 10 % ethanol turns out to be 3.46. That is, no flux takes place during rearrangement of the microfibril [9, 10].

Filtration Characteristics with Membrane Dispersion

Permeation experiments were conducted by submerging membrane modules in the bioreactor to investigate the fil-
Filtration characteristics with respect to the dispersion of HFMs. The transmembrane pressure (TMP) was measured via a suction gauge attached between the membrane modules and a suction pump. The permeate flux can be calculated from

\[
\text{Permeate Flux} (\text{L/m}^2 \cdot \text{hr}) = \frac{V}{A \cdot T}
\]  

(6)

where \(V\) is the filtrate volume (l), \(A\) is the area of the membrane (m²), and \(T\) is the filtration time (hour).

The filtrate flowrate was measured for the initial 15 min at a relatively low pressure; large differences appeared according to the type of membrane module. During the initial 15 min of the experiments, the average filtrate flowrates for 2, 4, and 10 bundles were 55.0, 63.0, and 75.5 mL/min, respectively. Thus, the influent flow rate was regulated differently for each module to maintain a constant effective reactor volume. Because of differences in the initial filtrate flowrates, the HRT of each reactor was ca. 10.4, 8.6, and 7.2 h, respectively.

The permeate flux (PF) and the transmembrane pressure (TMP) change are plotted against time at a suction pressure of 0.5 kgf/cm² in Figure 6. During the initial 11 h of the experiments, the average PF for each bundle was 15.0, 16.0, and 20.4 L/m², respectively. The TMPs were 31.8, 17.4, and 13.8 cmHg, respectively. Among the three bundles, the 10 bundle system exhibits the highest PF and the lowest TMP. As the permeation time increases, there is a trend of declining PF and increasing TMP. Figure 6 indicates that the change of TMP with operating time was due to the HFM dispersion. The increased space between each HFM and the lower degree of membrane conglomeration may reduce fouling between each HFM, resulting in lower permeation resistance [11,12].

Filtration Characteristics with Packing Density and Suction Pressure

The permeate flux (PF) and the transmembrane pressure (TMP) are plotted against the membrane packing density...
in Figures 7 and 8, respectively. We observe a trend of a lower packing density resulting in less membrane fouling, a high overall PF, and a low TMP. The steady state of the PF was achieved after 4 days. As shown in Figure 9, at the steady state the PF depends on the packing density (average values of 31.3 L/m$^2\cdot$h with 7.52~30.08 threads/cm$^2$, 20.7 L/m$^2\cdot$h with 45.11~60.15 threads/cm$^2$, and 32.7 L/m$^2\cdot$h with 30.08 threads/cm$^2$). Figures 10 and 11 show the PF and the TMP plotted against the suction pressure with a packing density of 30.08 threads/cm$^2$. When the suction pressure was 0.5 kgf/cm$^2$, the PF was high, but the TMP was low [13].

**Filtration Characteristics with MLSS Change**

With various MLSS concentrations in the aeration tank, the variations of the permeate flux and the transmembrane pressure were investigated and plotted against the packing density in Figure 12. The higher the MLSS concentration, the smaller the permeate flux due to increasing membrane fouling and the higher the transmembrane pressure.

**Optimum MLSS Concentration for Submerged MBR**

A batch-activated sludge reactor was used to obtain the yield coefficient (Y) and the endogenous decay coefficient (kd) to determine the optimum MLSS concentration for a submerged MBR. As seen in Figure 13, the confidence level for each coefficient, by linearization, is more than 95%. According to Equation (2), the slope is 0.6464, which corresponds to Y, and the intercept, kd, is 0.0712 day$^{-1}$.

Knowing the two coefficients, the appropriate organics loading can be calculated from Equation (4) to determine...
the optimum MLSS concentration for a long period of operation. Figure 14 indicates that an organics loading of 300 ∼ 600 mg COD/L · day is the most efficient to operate in the long term with MLSS concentrations ranging from 3800 to 8200 mg/L. This result indicates that if the membrane changing time is adequate and a suitable organic loading is provided, it is possible to operate the system for a long time without residue sludge production [14,15]. Also, we confirmed that long-term operation is possible with MLSS concentrations of 5000 ∼ 7000 mg/L and organic loadings of ca. 500 mg COD/L · day.

Conclusions

The permeate characteristics and the operating conditions for membrane bioreactors were investigated with dispersing hollow-fiber membranes. The results are summarized as follows:

1) With an increase of the ethanol (as solvent) concentration, the amount of eluted matter increased and the elution rate accelerated up to 70 % ethanol and then remained constant for a long period of operation. However, with a polar solvent concentration greater than 70 %, consistent membrane fouling and a lower SS removal efficiency were observed.

2) The initial permeate flux was stabilized at ethanol concentrations over 20 %. The optimal amount of ethanol in the solvent mixture was 20 ∼ 30 %.

3) By dispersing the hollow-fiber membranes, a high permeate flux (PF) was achieved at a low transmembrane pressure (TMP). The lower the membrane packing density, the higher the PF and the lower the TMP. The highest PF in this study was obtained with a packing density of 30.08 threads/cm².

4) When the packing density of HF decreased, the PF increased. The average values of PF were 31.3 and 20.7 L/m² · h in the range of packing densities of 7.52 ∼ 30.08 and 45.11 ∼ 60.15 threads/cm², respectively.

5) When the MLSS concentration increased in the reactor, the PF decreased and the TMP increased, indicating that the MLSS concentration affects directly the fouling formation. The 20-membrane module exhibited the most stable PF and the highest flux. The amount of PF decreased with an increase of the MLSS concentration.

6) From our experiments, the yield and decay coefficients were 0.6464 and 0.0712 day⁻¹, respectively. For long-term operation without residue sludge production, the optimum organics loading is ca. 300 ∼ 600 mg COD/L · day with an MLSS concentration of 3800 ∼ 8200 mg/L.

From these results, when activated sludge is directly used for solid/liquid separation, it is recommended that the system be operated at a lower transmembrane pressure by dispersing membranes to delay membrane fouling and to maintain a low permeation resistance. It is possible to develop submerged MBR systems to update wastewater treatment plants and gray water treatment systems.

References

2. I. Bemberis, P. J. Hubbard, and F. B. Leonard, Presented paper at Winter Meeting 1, American Soc. of Agri. Engineers (1971).