Environ. Eng. Res. Vol. 11, No. 2, pp. 91~98, 2006
Korean Society of Environmental Engineers

THE EFFECT OF AIR BUBBLES FROM DISSOLVED GASES ON THE MEMBRANE FOULING IN THE HOLLOW FIBER SUBMERGED MEMBRANE BIO-REACTOR (SMBR)

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(received December 2005, accepted April 2006)

Abstract : There is a possibility of the production of the air bubbles in membrane pores due to the reduction in pressure during membrane filtration. The effect of fine air bubbles from dissolved gases on microfiltration was investigated in the submerged membrane bio-reactor (SMBR). The R_{air} (air bubble resistance) was defined as the filtration resistance due to the air bubbles formed from the gasification of dissolved gases. From the results of filtration tests using pure water with changes in the dissolved oxygen concentration, the air bubbles from dissolved gases were confirmed to act as a foulant and; thus, increase the filtration resistance. The standard pore blocking and cake filtration models, SPBM and CFM, respectively, were applied to investigate the mechanism of air bubble fouling on a hollow fiber membrane. However, the application of the SPBM and CFM were limited in explaining the mechanism due to the properties of air bubble. With a simple comparison of the different filtration resistances, the R_{air} portion was below 1% of the total filtration resistance during sludge filtration. Therefore, the air bubbles from dissolved gases would only be a minor foulant in the SMBR. However, under the conditions of a high gasification rate from dissolved gases, the *effect of air bubble fouling should be considered in microfiltration.*

Key Words : Air bubble fouling, MBR, Hollow fiber membrane, Filtration resistance, Microfiltration

INTRODUCTION

The combination of membrane filtration for solid-liquid separation and biological activated sludge process, commonly known as a membrane bio-reactor (MBR), has gained considerable attention in wastewater treatment, reclamation and reuse. In this process, the membrane modules take the place of either the clarifier in a biological treatment process for suspended biomass or conventional separation devices to solve settling problems. It has many advantages, with respect to complete solids removal, significant physical disinfection capability, easiness for automation and small footprint requirement.[1-3] In addition, it also pro-

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duces less sludge than conventional activated sludge systems, due to the relatively low food to micro-organism ratio and long sludge retention time.[4)]

However, membrane fouling and the relatively high capital and operating costs are considered disadvantages of the MBR. Factors affecting membrane fouling in the MBR can be divided according to the membrane (configuration, mineral, hydrophobicity, pore size and porosity), biomass characteristics (Mixed Liquor Suspended Solids (MLSS), Extracellular Polymeric Substances (EPS), floc structure and dissolved matter) and operating conditions (aeration, hydraulic retention time (HRT), solids retention time (SRT), trans-membrane pressure (TMP) and cross-flow velocity).[5)] In terms of filtration resistance, the membrane (R_m), pore blocking (R_p) and cake (R_c) resistances are the main components of the total filtration resistance (R_t) in the MBR. Various fouling materials affect each of these resistances, increasing the total filtration resistance, and causing the permeate flux decline.

With the MBR, two main configurations are used in practice; one is the cross-flow MBR, with retentate circulation to the bioreactor. The other is the submerged MBR (SMBR), with the membrane filtration unit immerged directly in the bioreactor. Nowadays, many MBR applications use hollow fiber microfiltration with the SMBR.[6-9)]

In the aerobic SMBR, aeration is inevitable to supply oxygen for the aerobic biological reaction and remove sludge cake from the membrane surface. Some researchers have investigated the effect of the aeration intensity on the filtration performance in the SMBR.[10-12)] Usually, these have focused on the flux enhancement due to aeration in the SMBR. However, there is a possibility for the production of air bubbles in the membrane pores due to the reduction in pressure during membrane filtration. A hypothesis was suggested that gasified fine air bubbles, formed from dissolved gases when passing through the membrane pores, could foul the porous hollow fiber membrane.

The objective of this study is to investigate the effects of air bubbles in the hollow fiber membrane pores of the SMBR. The filtration tests, using pure water, were conducted using various dissolved oxygen concentration to verify air bubble fouling in hollow fiber microfiltration. Filtration models were applied to investigate the mechanism of air bubble fouling, with a critical flux test for the air bubble fouling also performed. Finally, the term "R_{air}" was introduced to signify the filtration resistance induced by air bubbles, which was compared with the filtration resistances from tests on various feeds (pure water, yeast, protein, mixture of yeast and protein, and sludge), to try and explain the significance of air bubbles on the fouling of hollow fiber membranes.

MATERIALS AND METHODS

Gravitational Hollow Fiber Filtration Apparatus

The schematic diagram of the hollow fiber microfiltration apparatus is shown in Figure 1. A 200 cm high plexiglass reactor (volume; 21 L) was used for the air bubble fouling experiment. The pressure could be controlled by the water level in the apparatus. Permeate was recycled, using a pump (Materflux® L/S™, Cole-Parmer Instrument Company, USA), to maintain a constant water level.

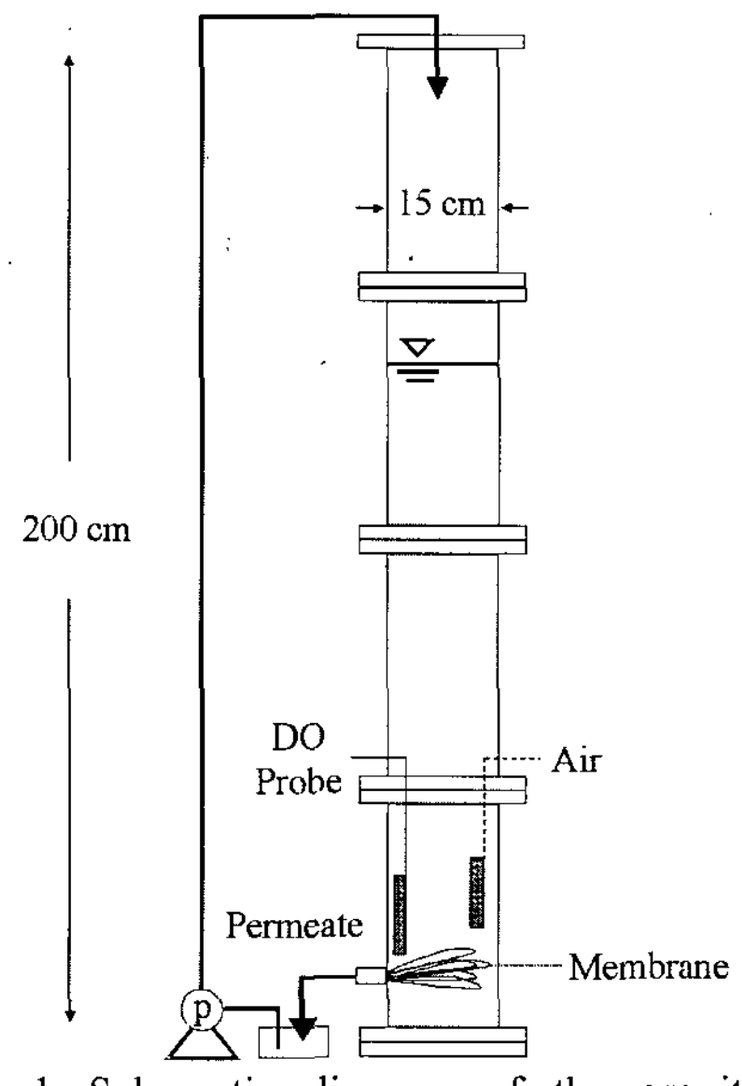

Figure 1. Schematic diagram of the gravitational filtration apparatus.

Table 1. The characteristics of the hollow fiber membrane

Items	Specifications
Membrane material	Polypropylene (PP)
Type of membrane	Hollow fiber
Pore size (μm)	0.4
Outer diameter (μm)	520
Inner diameter (μm)	360
Tube length (mm)	343
Surface area (cm^2)	77.4
Sealant	Epoxy

The membrane characteristics are presented in Table 1. A hollow fiber membrane was equipped at the bottom part of the reactor. The membrane was potted with epoxy sealant and soaked in distilled water for at least 24 hours prior to each experiment. The potted membrane was easily removed from the reactor at the silicon cab connection. In the model applications and dissolved oxygen tests, the air, at a rate of 2.5 L/min, was supplied above the membrane to avoid direct contact of the membrane surface with air bubbles. The dissolved oxygen concentration was measured using a DO meter (Thermo Orion model 810, Orion Research Inc., USA) immediately after turning off the aeration on the opposite side to the aerator to prevent the direct contact with the air bubbles.

Suctional Hollow Fiber Filtration Apparatus

Figure 2 shows the suctional microfiltration apparatus for the critical flux test with permeate recycling. Permeate was recycled using a circulation pump (Materflux® L/S™, Cole-Parmer Instrument Company, USA). A pump drive, equipped with a pump head (Micropump, Inc., USA), was used to create suction pressure within the range 0.0~0.6 bar, which was measured using a pressure gauge.

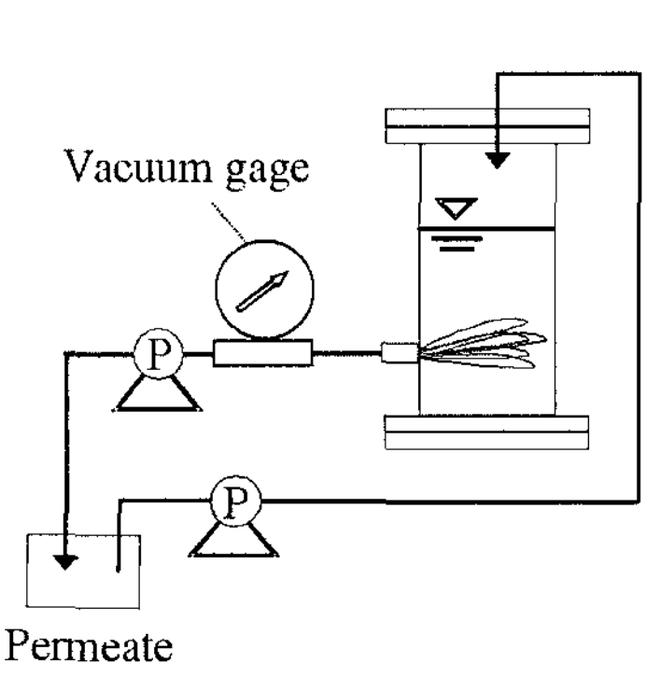

Figure 2. Schematic diagram of the suctional filtration apparatus for the critical flux test.

Filtration Test with Various Feeds

Yeast (*Saccharomyces cerevisiae*) suspension was used to represent large particle in the feed. PBS (Phosphate Buffer Solution; 5mM NaH_2PO_4; 5mM $Na_2HPO_4 \cdot 12H_2O$, 0.15M NaCl, 0.01% NaN_3) was used to prevent yeast cell lysis, and to ensure the yeast was a pure form, the sample of yeast was treated as follows: washed three times in the PBS, centrifuged at 2,500 rpm for 15 min and each supernatant discarded. The concentration was expressed in dry weight per liter of suspension. To represent feed containing macromolecules, a protein (Albumin, Bovine Initial fractionation by heat shock fraction V, 66,430Da) was purchased from Sigma chemical company and dissolved in PBS. Sludge was taken from the membrane bio-reactor (MBR), operating with a solids retention time (SRT), hydraulic retention time (HRT) and mixed liquor suspended solids (MLSS) of 30 days, 18 hours and 5,250 mg/L, respectively. The pH of the sludge was 7.2. In the tests with the various feeds, the air, at a rate of 2.5 L/min, was used to mix the feed. Filtration tests were conducted with the various feeds at a pressure of 0.1 bar and temperature of 25°C.

THEORY

Gasification of Dissolved Gases from the Feed

The imposed pressure drop is the driving force of permeate production during microfiltration. The pressure on the feed side is higher than that of the permeate in the SMBR, which can induce gasification of dissolved gases during filtration. The solubility of a gas is decreased due to a reduction in pressure at constant temperature. The relationship between pressure and the concentration of dissolved gas is given by Henry's law:

$$C = k_H P \quad (1)$$

Table 2. The theoretical dissolved gases in water at 1.013 bar and 25°C

Items	Nitrogen	Oxygen	Carbon dioxide
Molecular weight (g)	28.0	32.0	44.0
Henry's law constant (k_H; mol/L·bar)	6.42E-4	1.28E-3	3.36E-2
Partial pressure (bar)	0.781	0.209	0.003
Dissolved molarity (mol/L)	5.01E-4	2.69E-4	1.01E-4
Dissolved concentration (mg/L)	14.0	8.6	4.4
Dissolved volume (mL/L)	12.3	6.6	2.5

Where, P=partial pressure of the gaseous solute (bar), C=molarity of the dissolved gas (mol/L), k_H=Henry's law constant (mol/L·bar).

Table 2 shows the concentrations and volumes of theoretical dissolved gases at 25°C and 1.013 bar. The assumptions are following;

1. The system is open and in an equilibrium.
2. Air consists of nitrogen, oxygen and carbon dioxide, which are ideal gas.
3. Dissolved gases are saturated in the feed.
4. There are no reactions in the solution (e.g. ignore carbonate system).

Usually, the pressure applied in the SMBR is within the range 0.0-0.6 bar. For example, if the pressure difference is 0.6 bar, the theoretical total gases produced per 1 liter of permeate is 12.8 mL on passing through the membrane pores at 25°C. The volume of dissolved gases that become gasified increases with increasing pressure drop.

Filtration Model

Resistance-in-series model

Darcy's Law states that the flux is directly proportional to the potential pressure drop and inversely proportional to the resistance. For porous membrane systems, the flux can be expressed by the resistance-in-series model given below:

$$J=\frac{\Delta P}{R_t\cdot\mu} \tag{2}$$

$$R_t=R_m+R_c+R_p+R_{air} \tag{3}$$

Where, J=permeate flux (m/s), ΔP=applied pressure (Pa), μ=viscosity of feed solution (Pa·s), R_t=total hydraulic resistance (1/m), R_m=membrane resistance for pure water (1/m), R_c=resistance caused by cake layer (1/m), R_p=resistance caused by pore blocking (1/m) and R_{air}=air bubble resistance from dissolved gases (1/m).

Standard pore blocking model (SPBM)

Hermia[13] summarized different filtration models that could be applied for non-Newtonian fluids and constant pressure filtration. These can be expressed as simple equations relating the filtrate flow (Q; m^3/s), permeate volume (V; m^3) and time (t; s).[14]

When the particle diameter is much less than that of the membrane pores, the particles will pass through the pores. The particles will deposit on the pore walls, subsequently reducing the pore diameter and; thus, the pore volume. This can be expressed by the following equation:

$$\frac{t}{V}=\frac{K_s}{2}t+\frac{1}{Q_0} \tag{4}$$

Where, Q_o is the initial permeate flux and K_s ($1/m^3$) the filtration constants in the SPBM.

Cake Filtration Model (CFM)

In the case of large particles, which are mostly unable to enter through the pores, a deposit will develop in the form of a cake on the membrane surface. This can be expressed by the following equation:

$$\frac{t}{V}=\frac{K_c}{2}V+\frac{1}{Q_0} \tag{5}$$

Where, Q_o is the initial permeate flux and K_c (s/m^6) the filtration constant in the CFM.

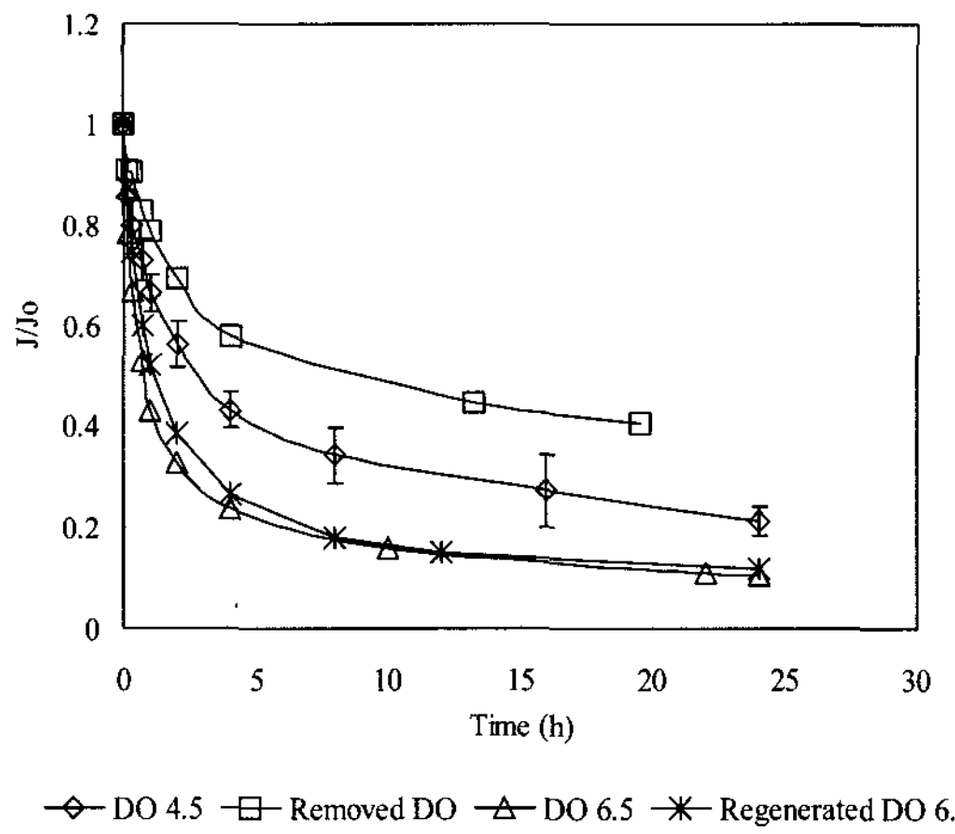

Figure 3. Air bubble fouling with pure water at various dissolved oxygen concentrations. (Removed DO=DO 0.0 mg/L, DO 4.5=DO 4.5 mg/L, DO 6.5=DO 6.5 mg/L, regenerated DO 6.5=filtration under DO 6.5 mg/L after the sonication)

RESULTS

Air Bubble Fouling

The changes in the normalized flux were measured in terms of the dissolved oxygen (DO) concentration to investigate the effect of air bubble fouling in microfiltration. Pure water, 18 MΩ, was used at a pressure of 0.1 bar, as determined by the water height. Figure 3 shows the results of the air bubble fouling. Although pure water was used, the graphs obtained with the various DO concentrations revealed flux declines as shown in Figure 3. By turning the aeration on and off the DO concentrations was maintained at 6.5 and 4.5 mg/L for the DO 6.5 and 4.5 tests, respectively. The DO was removed by the addition of 1 g/L of Na_2SO_3 (removed DO test), but this recovered after 5 hours. When a higher DO concentration was applied, a higher rate of flux decline was observed. The reason for the flux decline in the removed DO test was assumed to be due to the effect of other gases, such as nitrogen and carbon dioxide. From the data shown in Table 2, oxygen accounted for about 31% of the total dissolved gases.

After the DO 6.5 test, the fouled membrane was removed from the apparatus, and sonication (Ultrasonic cleaner, Model FS21H, Fisher Scientific, USA) applied for 1 minute to remove the air bubbles trapped in the test membrane, with the production of fine air bubbles observed from the membrane pores. After the sonication, the regenerated membrane was used to conduct further filtration tests (regenerated DO 6.5 test), with the regenerated membrane showing a similar trend in the DO 6.5 test as the original membrane. When a different membrane material (polyethylene, pore size 0.4 μm) was used with aeration, the flux decline in the DO 6.5 test was observed to be similar to the other material. Therefore, these results indicated that air bubbles were produced during the filtration, which played a role as a foulant.

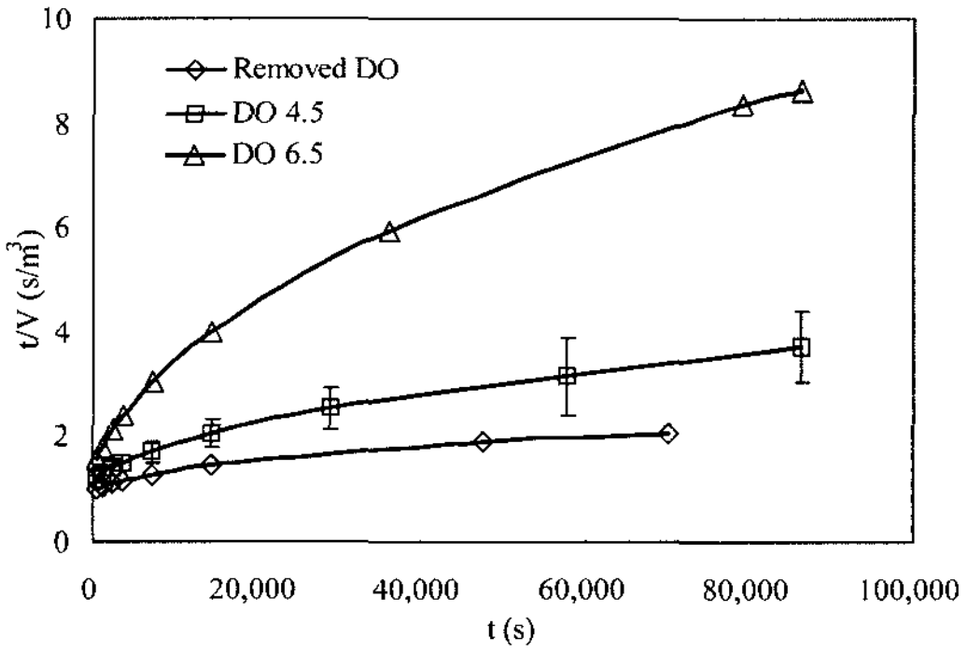

Figure 4. Application of the SPBM to air bubble fouling.

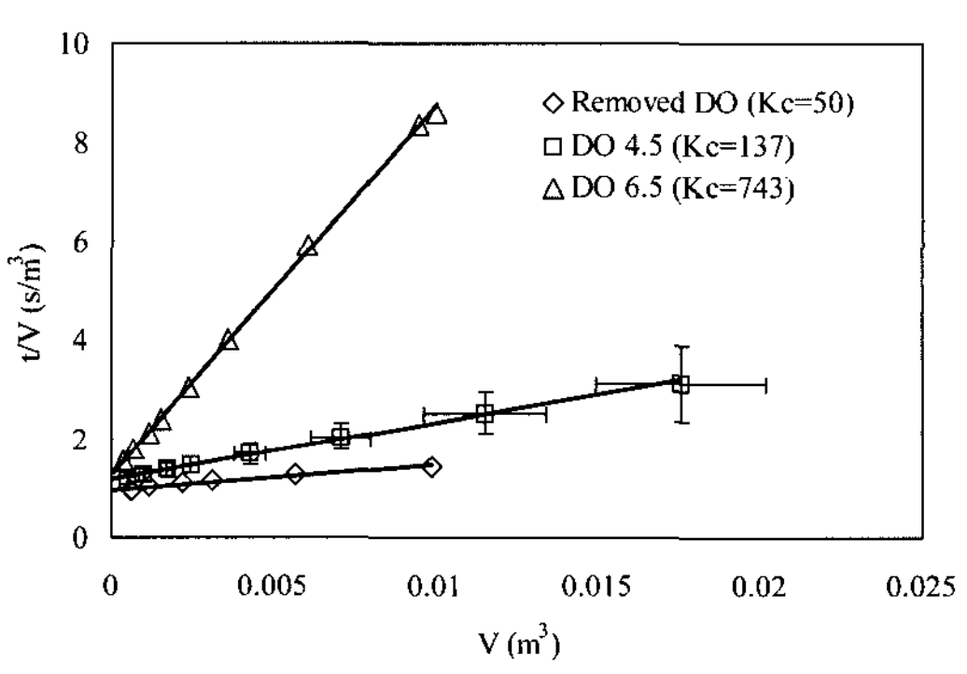

Figure 5. Application of the CFM to air bubble fouling.

Mechanism of Air Bubble Fouling

The SPBM and CFM were used to help discover the mechanism of air bubble fouling. Figures 4 and 5 shows the application of the SPBM and CFM, respectively, at each DO concentration. The plot of t/V vs. t was not linear in the application of the SPBM, while that of t/V vs. V showed a linear relationship in the CFM, which meant the air bubble fouling followed that of the CFM. From the theory of CFM, the result implies that the air bubbles too large to enter the pore form a cake of air bubbles on the membrane surface. However, the production of air bubbles was not observed on the membrane surface during the pure water filtration test. It was assumed that there were limitations in applying these models, as air bubbles are not rigid particles and can easily detached from the membrane pores due to the permeate flow.

Critical Flux of Air Bubble Fouling

The critical flux for "R_{air}" was measured so that under the condition of a DO concentration of 4.5 mg/L the air bubble fouling could be ignored. The critical flux is defined as some specific value where fouling is not observed when the flux is maintained.[15,16] Figure 6 shows the critical flux measurement with pure water filtration. Flux 1 showed the flux changes with continuous suction. The flux was measured every 1 minute as the suction pressure was increased by the suction pump step by step. Flux 2 was a plot of the initial flux at each pressure, with membrane changes where the slope of the line was linear

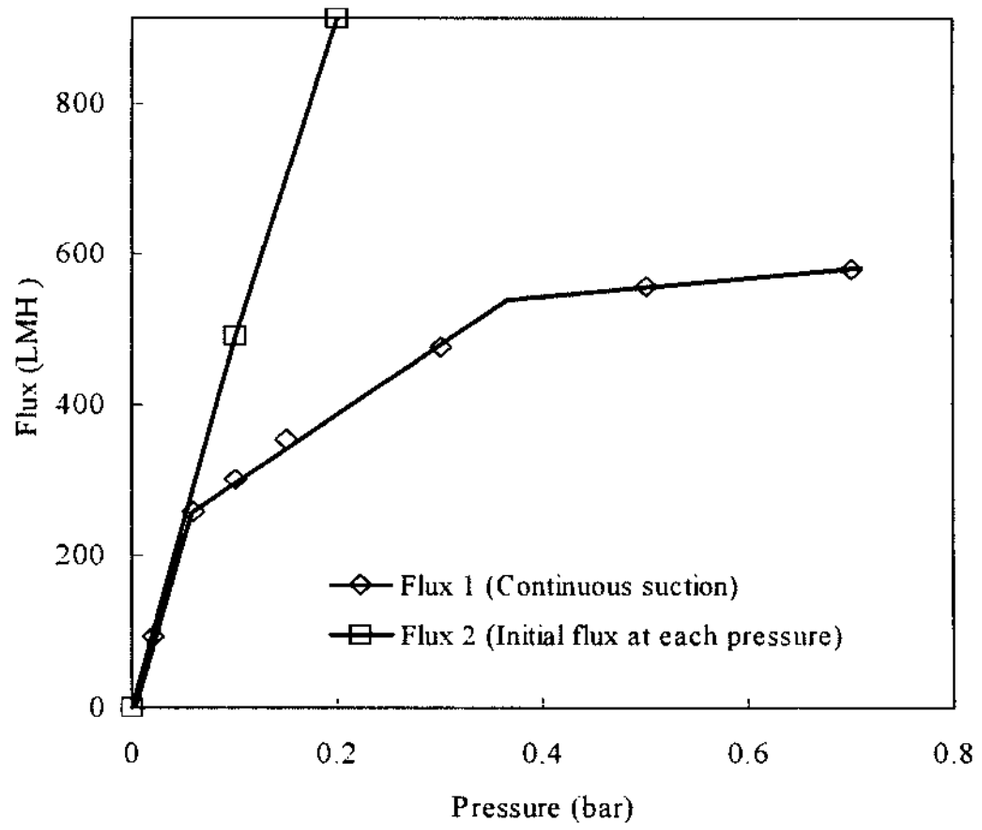

Figure 6. The critical flux measurement for air bubble fouling.

in the pressure range 0.0-0.6 bar. From the results of permeate fluxes 1 and 2, around 250 $L/m^2 \cdot hr$ (LMH) was found as the critical flux for air bubble fouling. There was a sudden flux decline above a pressure of 0.4 bar, where fine air bubbles were observed on the membrane surface and in the suction tube.

Comparison Tests

In order to analyze the portion of air bubble resistance in the filtration of various feeds, the membrane (R_m), pore blocking (R_p), cake (R_c) and air bubble (R_{air}) resistances were calculated, as shown in the Table 3. Although the fouling rate should fluctuate in the tests with various feeds, the air bubble resistance was fixed at that for the pure water test at a DO concentration of 4.5 mg/L for a simple comparison of the resistance

Table 3. Filtration resistances with various feeds after 2 hours of filtration (unit: 10^{10} m^{-1})

Feed	R_m (%)	R_p (%)	R_c (%)	R_{air} (%)	R_t (%)
Pure water (DO 4.5 mg/L)	6.53 (83.1)	-	-	1.33 (16.9)	7.86 (100)
Yeast 1.0 g/L	6.53 (14.9)	-	35.84 (82.1)	1.33 (3.0)	43.7 (100)
Yeast 2.0 g/L	6.53 (11.3)	-	49.94 (86.4)	1.33 (2.3)	57.8 (100)
Protein 0.5 g/L	6.53 (34.2)	11.24 (59.1)	-	1.33 (6.7)	19.1 (100)
Yeast 1.0 g/L + Protein 0.5 g/L	6.53 (11.6)	48.54 (86)		1.33 (2.4)	56.4 (100)
Yeast 2.0 g/L + Protein 0.5 g/L	6.53 (7.8)	75.74 (90.6)		1.33 (1.6)	83.6 (100)
Sludge 5.25 g/L	6.53 (3.3)	190.14 (96.03)		1.33 (0.67)	198 (100)

values through equation (1). In this study, gas reactions in the solution were ignored and theoretical calculations were conducted assuming ideal conditions. The test with the highest concentration of sludge showed the most severe fouling. The mixture of yeast and protein showed a higher flux decline rate than the tests with yeast alone. The "R_{air}" portions of "R_t" were below 5%, with the exception of the pure water and protein tests. As the concentration of yeast was increased, the portion of "R_c" also increased, but the portion of "R_{air}" decreased. In the test with the mixture of yeast and protein, the "$R_p + R_c$" was higher than the sum of the resistances in each of the tests for yeast and protein alone. It was assumed that the pores of the yeast cake, as well as those of the membrane pores, were blocked by protein. The "R_{air}" portion in the sludge test was below 1% of the "R_t".

DISCUSSIONS

From the graphs obtained for the filtration tests with pure water, flux declines due to the air bubble fouling were observed. However, the resistance due to air bubble fouling should be minor comparing to other total filtration resistances in the SMBR. The "R_{air}" portion in the sludge test was below 1% of the "R_t". Therefore, the effect of air bubble fouling on microfiltration should only be a minor foulant in the SMBR. The trapped air could also pass through the hollow fiber membrane during air scouring in the SMBR operation. However, under conditions of high dissolved gas concentrations and reduction in pressure, air bubble fouling should be considered in microfiltration.

The model application could not explain the air bubble fouling. The air bubbles could not be considered as colloidal materials or rigid particles. In the initial stage of the filtration, fine air bubbles would be produced due to the reduction in pressure and then attach to the wall of membrane pores. During the continuous filtration, it was assumed that the fine air bubbles in the membrane pores could combine to form larger bubbles, with some becoming trapped in the pore or moving to the permeate side. Therefore, further research will be required to explain the mechanism of air bubble fouling using a corrected model application or by direct observation.

Air bubble fouling could depend on the aeration conditions and membrane characteristics. Further research will be needed to investigate the effect of air bubble fouling with different membrane pore sizes, aeration rates and degrees of aeration.

CONCLUSIONS

In the filtration experiments for the effect of gasified air bubbles from dissolved gases on the hollow fiber microfiltration, the following results were obtained.

1) From the flux decline of pure water with the various dissolved oxygen concentrations, the air bubbles from the dissolved gases could play a role as a foulant, causing an increase in the filtration resistance.
2) The application of the SPBM and CFM were limited in their explanation of the mechanism of air bubble fouling due to the properties of air bubble.
3) The critical flux for the air bubble fouling was around 250 LMH, where the air bubble fouling could be ignored if the flux was maintained.
4) The effect of air bubble fouling on the hollow fiber microfiltration membrane was minor in the SMBR. However, under conditions of high dissolved gases concentration and reduction in pressure, air bubble fouling should be considered in microfiltration.

ACKNOWLEDGEMENTS

This research was supported by the grant (4-1-2) from the Sustainable Water Resources Research Center (SWRRC) of 21st century frontier R&D program and, in part, by the Center for Distributed Sensor Network at Gwangju Institute of Science and Technology (GIST).

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