공기주입 노즐관이 장착된 관형막의 투과특성

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Permeation Characteristics of the Tubular Membrane Module Equipped wtih the Air Injection Nozzle Tube

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요   약: 분리막 오염을 감소시키고 투과유속 향상을 위하여 관형분리막 모듈 내에 공기 분사노즐관을 삽입시켰다. 분리막의 평균 기공크기는 0.1 μm이며 이스트를 오염물질로 사용하였다. 모든 투과실험은 노즐관을 모듈에 장착하고 공기를 주입하지 않는 실험을 먼저 실시하고 연속해서 공기를 주입하는 투과실험을 하였다. 그 다음 노즐관을 제거한 후 공기를 주입시키지 않으면서 투과유속을 측정하였다. 측정된 투과유속은 공기주입 효과를 분석하기 위하여 비교하였다. 공기주입에 대한 투과유속은 거의 일정하거나 증가하였다. 노즐관이 장착되고 공기 주입을 하지 않을 경우의 투과유속이 빈 관형 모듈의 경우보다 높았다. 운전압력을 0.4 bar까지 감소시키면 노즐관이 장착되지 않는 경우와 비교하여 공기를 주입할 경우 투과유속이 21%까지 향상되었다. 기체량이 증가하여 기/액체 2상 흐름이 stratified-smooth에서 intermittent 상태로 변화됨에 따라서 공기 주입에 의한 투과유속은 30% 이상으로 증가하였다.

Abstract: The air injection nozzle tube was inserted inside of the tubular membrane module to reduce membrane fouling and improve the permeate flux. The average pore size of membrane was 0.1 μm and the yeast was used as a foulant. All of permeate experiments were started without air injection for the module equipped with the nozzle tube, then carried out continuously with air injection. Finally, the nozzle tube was removed from the module and the permeate was measured without air injection. The measured permeate fluxes were compared to examine the effect of air injection. The fluxes for air injection were consistently maintained or increased. The fluxes of no-air injection with the nozzle tube were greater than those of the empty tubular module. As operating pressure decreased to 0.4 bar, the flux enhancement of air injection based on no-nozzle case increased to 21%. Flux enhancements of air injection were above 30% as the gas/liquid two-phase flow was changed from the stratified-smooth to the intermittent pattern due to increase of gas flowrate.

Keywords: injection nozzles, air injection, yeast solution, tubular membrane, permeate flux, fouling

1. Introduction

Development of the high-performance materials and/or effective operational methods to reduce or control membrane fouling is the main membrane research area during last few decades[1-3]. Some methods have

been proposed the inserted tube[4-5], pulsatile flow[6], corrugated membrane surface[7], backwash using air to reduce membrane fouling and concentration polarization. Recently, many papers have pointed out that membrane fouling can be mitigated by using two phase gas-liquid flow in a horizontal tube. This method may be effec-

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(1) Feed tank
(2) Masterflex pump
(3) Membrane module
(4) Pressure gauge
(5) Pressure regulating valve
(6) Water flowmeter
(7) Air compressor

(8) Pressure regulating valve
(9) Air flowmeter
(10) Check valve
(11) Electronic balance
(12) Computer
(13) Stirrer
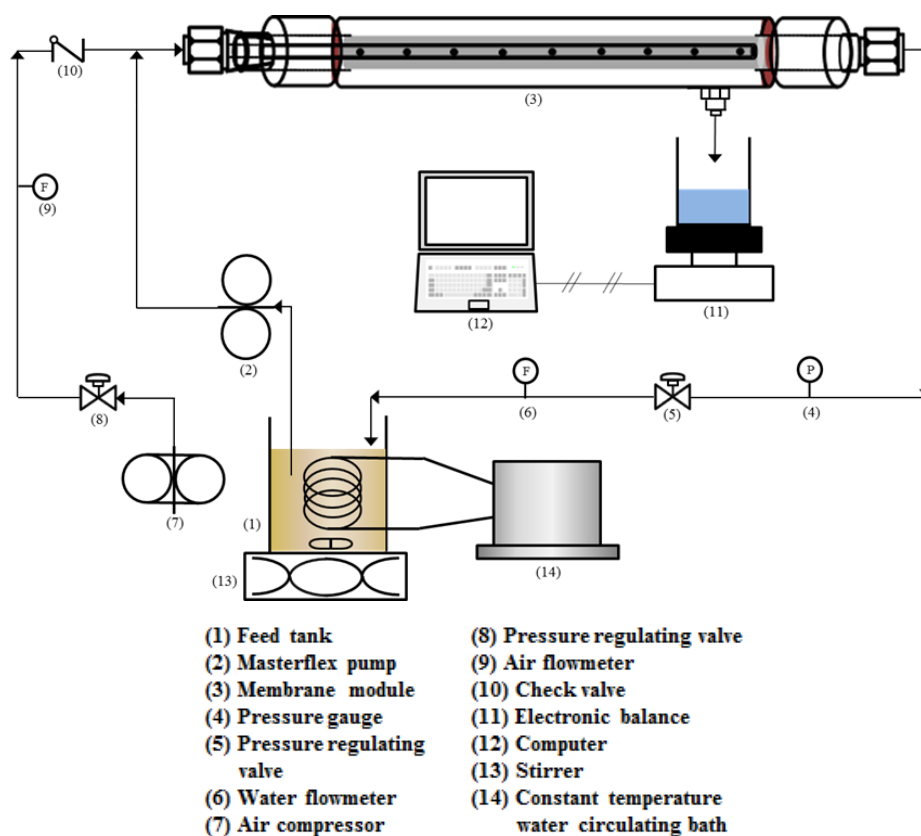(14) Constant temperature water circulating bath

Fig. 1. Schematic flow diagram of tubular membrane system[17].

tively controlled by shear stress or vortex generation by air injection on the membrane surface in most conditions[8-10]. The low gas flow in the slug region of two phase gas-liquid flow could be applied effectively to remove the particles near the membrane surface and improve permeate flux[11]. The slug flow is gas flow as large bullet shaped bubbles approaching the tube diameter of the tube. There may be followed small bubbles going along the slug[12]. Air injection in tubular membrane applications has proven to be an effective, cost-saving enhancement technique, however, influence of two phase gas-liquid flow needs to be investigated[13-14]. The back-flushing with conventional air injection is the way to reduce membrane fouling as well as improve permeate flux. The flux enhancement of cross-flow microfiltration membrane has been studied by Mikulášek[15], who has performed experiment of two techniques by injected air and back-flushing inside the membrane. The study was shown both of gas sparging and back-flushing method were effective in fouling problem but back-flushing was much more effect on membrane defouling when the back-pulse duration was shorter. Also, Fadaei[16] reported comparative assessment of the efficiencies of gas sparging and back-flushing for feed concentration. Gas sparging was shown greater efficiency in flux enhancement at higher concentration and back-flushing was more effective internal fouling due to pore blockage at lower feed concentration.

The objective of this study was determined the optimal operating conditions to reduce effectively membrane fouling of the membrane surface for permeate flux enhancement by air injection directly on the surface of the membrane. The nozzle was self-designed and equipped into the ceramic membrane of tubular module to inject the air[17].
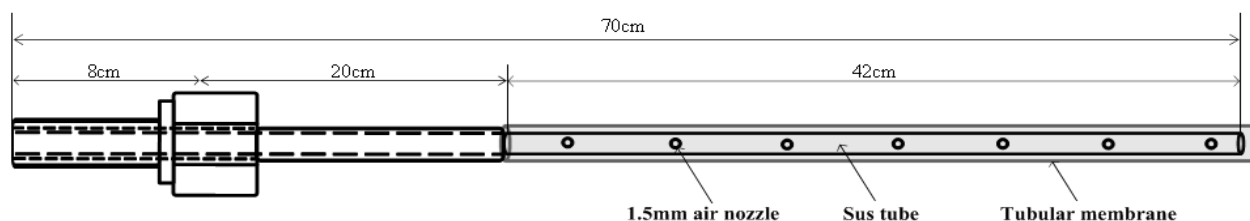
Fig. 2. Details of the air injection tube and nozzles.

2. Experimental

2.1. Experimental system

The experimental apparatus used in this study is shown schematically in Fig. 1. The system consisted of four parts: liquid, gas supplying sections, membrane module and permeate collecting section. The liquid feed was supplied from a 5 L plastic reservoir tank (1) to the self-designed transparent acrylic plastic tubular module (3) by the masterflex pump (2). The outlet pressure (4) of the module was measured and regulated by the valve (5). The feed flowrate was measured with a flowmeter (6) before the stream was recycled to the tank (1). The air was supplied to the membrane module (3) by the air compressor (7), regulated by the valve(8). The air flowrate was measured with an air flowmeter (9). The check valve (10) was installed in the air line to prevent the back flow of the liquid solution. The liquid and air feed lines were combined before the membrane module. Therefore gas-liquid two phase fluid was fed into the membrane module to reduce membrane fouling or concentration polarization. The permeate stream was collected in a permeate reservoir onto the electronic balance (11), and then periodically recycled to the feed tank in order to maintain an essentially constant feed concentration. The electronic balance interfaced to a computer (12) to collect permeate flux date. The particles in the feed tank were mixed well with the magnetic stirrer (13). The temperature of the feed solution was controlled by a cooling coil placed in the feed tank with the constant temperature circulating water bath (14).

2.1.1. Air injection nozzle tube

The self-designed air nozzle tube can be inserted inside of the tubular membrane module to reduce membrane fouling and improve the flux. The air nozzles of 1.5 mm diameter are placed every 2 cm outside the tube. The entire length of the air injection nozzle tube is inserted into the 42 cm tubular membrane module as shown in Fig. 2.

2.1.2. Experimental Materials

The experiments were performed using microfiltration, symmetric, slip casting, ceramic membrane (Nanopore Materials Co., Korea). The tubular membrane of 0.1 μm average pore size is the internal diameter of 1.2 cm, a length of 42 cm and an effective membrane area of 158 cm^2.

Pure water was used as permeate of tap water that passed through by pre-treatment, ultrafiltration and reverse osmosis membrane.

The particles were prepared the instant yeast which is containing yeast of 98.5% and manufactured by Jenico Co. The concentration of yeast was prepared with 0.01, 0.05, 0.1 and 0.2 wt% different concentrations of each 4 L for feed solution. The 0.5 N NaOH(Daejung, extra pure reagent) solution was used to remove the yeast from the tubular membrane.

2.2. Experimental procedures
2.2.1. Permeation experiments

The nozzle tube was equipped in the module and injected the air into the yeast solution. The yeast solution were prepared to using a stirrer to dissolve completely. All of experiments were started without air injection, then were carried out continuously with air
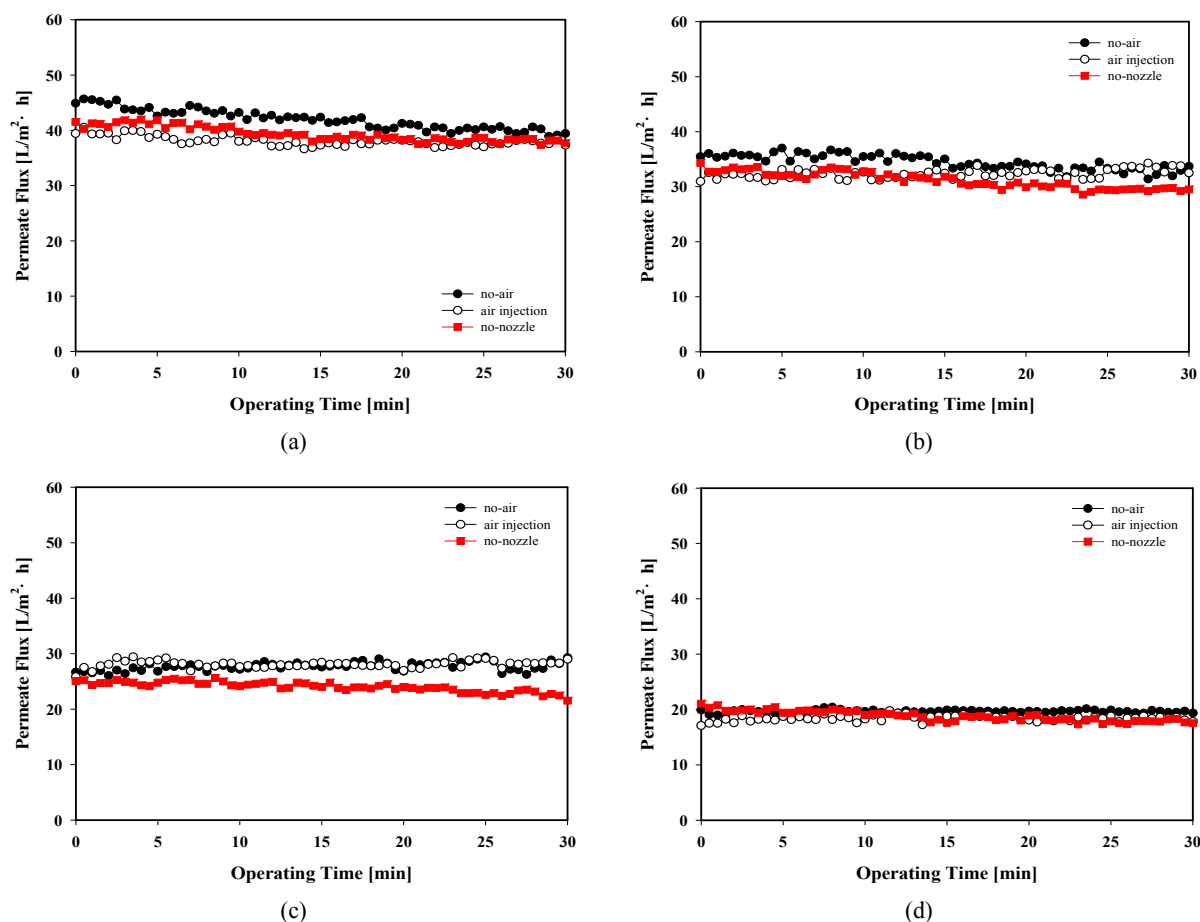
(a)

(b)

(c)

(d)

Fig. 3. Permeate fluxes as a function of operating pressure for 0.1 wt% yeast solution with 0.1 μm tubular membrane at 20°C and (a) 1.0 bar, (b) 0.8 bar, (c) 0.6 bar and (d) 0.4 bar.

injection. Finally, the nozzle was removed from the module and the permeate was measured without air injection. The permeate fluxes were measured with the variation of operating pressure, yeast concentration, gas and liquid flowrates. The temperature was fixed 20℃ and operating pressure can be able to regulate in the range of 0.1 bar because of vibration caused by two-phase gas and liquid. Operating pressure was controlled from 0.4 to 1.0 bar (every 0.2 bar), fixed gas flowrate of 2 L/min, liquid flowrate of 0.7 L/min and 0.1 wt% yeast solution for 30 min. The gas flowrate was controlled between 1 and 4 L/min, and liquid flowrate were 0.5, 0.7, 1.0 and 1.2 L/min within the allowable experimental range.

2.2.2. Membrane cleaning

After the permeation experiment, the physical and chemical cleaning were used in order to clean the fouled membrane. The tubular membrane surface was cleaned by flushing the pure water. After then, the tubular membrane was immersed in the 0.5 N NaOH solution for 4 hours to dissolve the yeast[18]. Finally, the membrane module was washed thoroughly and immersed in pure water until to carry out the following experiment.

3. Results and Discussion

3.1. Pressure effect on flux enhancement

The permeate flux (J) of 0.1 wt% yeast solution with respect to the operating pressure are as shown in Fig. 3. The other operating condition was fixed at 0.7

Table 1. Normalized Permeate Fluxes (J/J_0) and Flux Enhancement for Yeast Solution for 0.1 μm Tubular Membrane at Various Pressure

	Operating pressure	0.4 bar	0.6 bar	0.8 bar	1.0 bar
	no-air	1.03	1.06	0.91	0.87
J/J_0	air injection	1.03	1.06	1.06	0.98
	no-nozzle	0.85	0.92	0.90	0.92
Flux enhancement, %	air injection/no-air	0	0	16	13
	air injection/no-nozzle	21	15	18	7
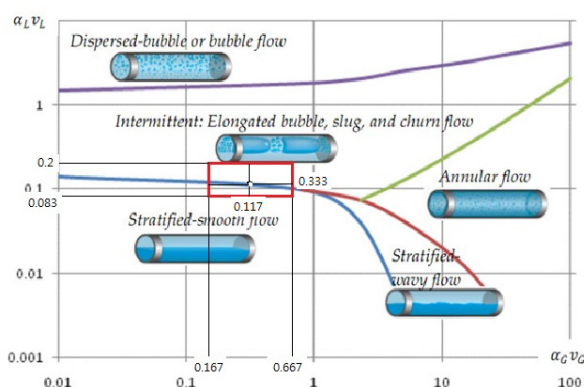
Fig. 4. Flow regime map in the tubular module from 1 and 0.5 to 4 and 1.2 L/min (or 0.167 and 0.083 to 0.667 and 0.2 m/s) of gas and liquid flowrates, respectively[19].

L/min liquid and 2 L/min gas flowrates. This liquid and gas two-phase flow was corresponded to the border of the stratified-smooth and the intermittent: elongated bubble, slug and churn flow condition at the end of injection nozzle as shown in Fig. 4[19]. Also, the operating pressure was fluctuated within 0.1 bar due to two phase flows when the air was injected into the module. The permeate fluxes of no-air injection decreased with respect to operating time. The permeate fluxes for no-air injection with the air injection tube in tubular module (so-called no-air) and empty tubular module without the air injection tube nozzle (so-called no-nozzle) cases continuously decreased from 45.2 to 39.1 LMH and from 41.2 to 38.1 LMH at 1 bar, respectively as shown in Fig. 3(a). In the case of no-nozzle, permeate fluxes were lower than the case of no-air. The air injection tube in the tubular membrane could induce flow eddies or flow mixing, then reduce membrane fouling. The permeate fluxes for air injection were consistently observed to maintain or increase. For instance, the permeate flux for air injection was maintained about 39 LMH at 1 bar without a significant change overall. The permeate fluxes decreased as the operating pressure decreased, and eventually the difference between the operational modes of no-air, air injection and no-nozzle was a little at 0.4 bar.

The normalized permeate flux ratios (J/J_0) based on the initial permeate flux (J_0) and flux enhancement for the operating pressure are summarized in Table 1 in order to evaluate the effect of air injection. The permeate flux due to air injection comparing no-air operation was not enhanced below 0.6 bar, but the flux enhancement was more than 13% above 0.8 bar. The flux enhancement due to air injection comparing no-nozzle operation was 21% at 0.4 bar, but decreased as the operating pressure increased. At lower operating pressure, the effect of flow eddies or mixing due to hindrance of the injection tube was greater than that of air injection. The eddies might be enough to reduce the fouling on the membrane surface. As operating pressure increased, the fouling layer could be more compactible, and the strength of the eddies might not be enough. However, air injection on the membrane surface through the air nozzle was observed relatively effective.

3.2. Concentration effect on flux enhancement

The permeate fluxes for yeast solution concentration are as shown in Fig. 5. The other operating condition was fixed at 0.8 bar, 0.7 L/min liquid and 2 L/min gas flowrates. The permeate fluxes were maintained with-

Table 2. Normalized Permeate Fluxes (J/J₀) and Flux Enhancement for Yeast Solution for 0.1 μm Tubular Membrane at Various Concentration

|	Yeast concentration	0.01 wt%	0.05 wt%	0.1 wt%	0.2 wt%
J/J_0	no-air	0.93	0.95	0.91	0.86
	air injection	1.06	1.05	1.09	1.03
	no-nozzle	0.96	0.89	0.86	0.85
Flux enhancement, %	air injection/no-air	14	11	20	20
	air injection/no-nozzle	10	18	27	21
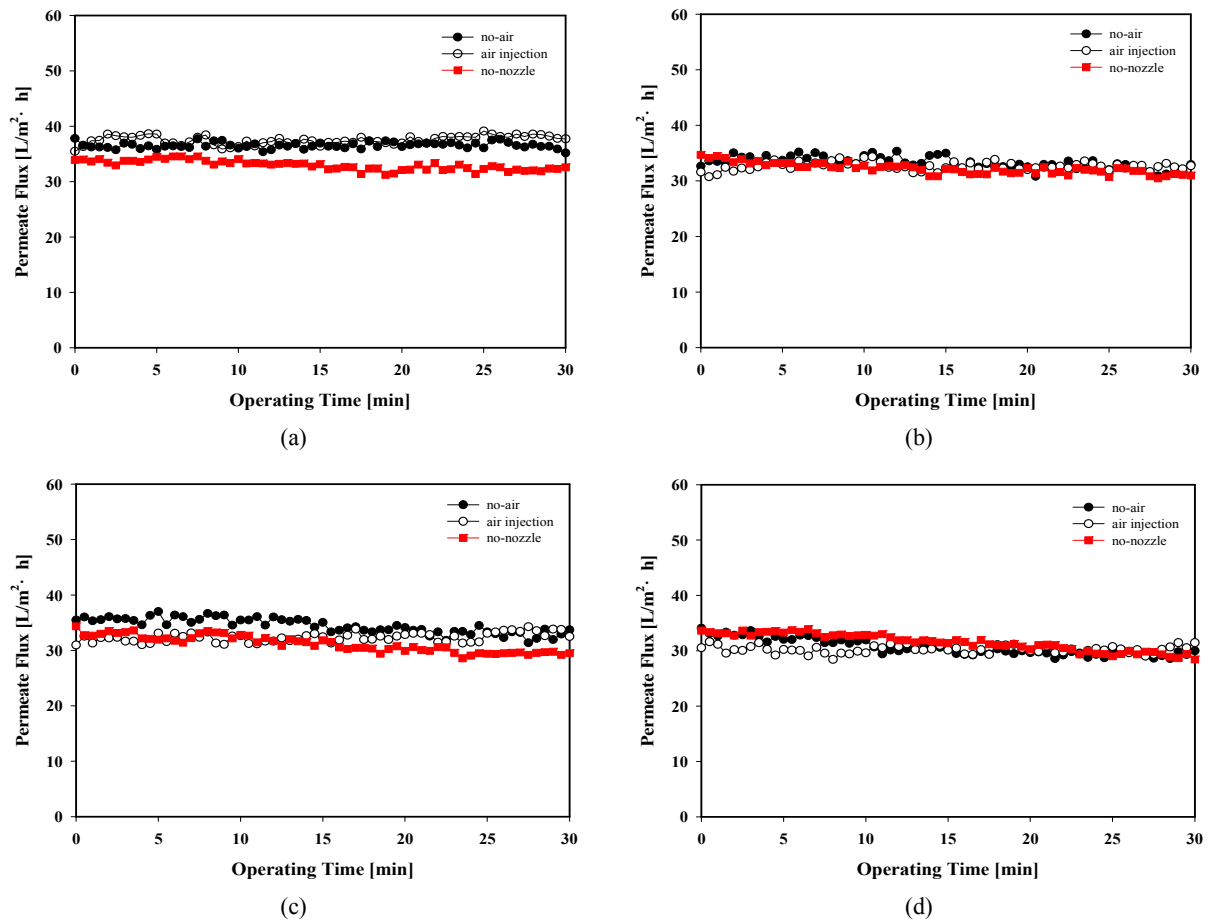
Fig. 5. Permeate fluxes as a function of concentration for yeast solution with 0.1 μm tubular membrane at 0.8 bar, 20°C and (a) 0.01 wt%, (b) 0.05 wt%, (c) 0.1 wt% and (d) 0.2 wt%.

out a significant change compared to the previous experiments for gas flowrate. The permeate fluxes for no-air and no-nozzle cases continuously decreased from 37.7 to 35.1 LMH and from 33.9 to 32.5 LMH at concentration of 0.01 wt%, respectively as shown in Fig. 5(a). Permeate fluxes for air injection case increased

from 35.4 to 37.7 LMH. The permeate fluxes decreased slightly as the concentration of yeast solution increased. However, permeate fluxes for air injection case increased from 31.5, 30.9 and 30.5 to 33.1, 33.7 and 31.5 LMH for 0.05, 0.1 and 0.2 wt% yeast solutions, respectively. This might be the reason why the

Table 3. Normalized Permeate Fluxes (J/J_0) and Flux Enhancement for Yeast Solution for 0.1 μm Tubular Membrane at Various Gas Flowrate

	Gas flowrate	1 L/min	2 L/min	3 L/min	4 L/min
J/J_0	no-air	0.85	0.91	0.85	0.86
	air injection	1.01	1.09	1.16	1.13
	no-nozzle	0.86	0.86	0.86	0.86
Flux enhancement, %	air injection/no-air	19	20	36	31
	air injection/no-nozzle	17	27	35	31
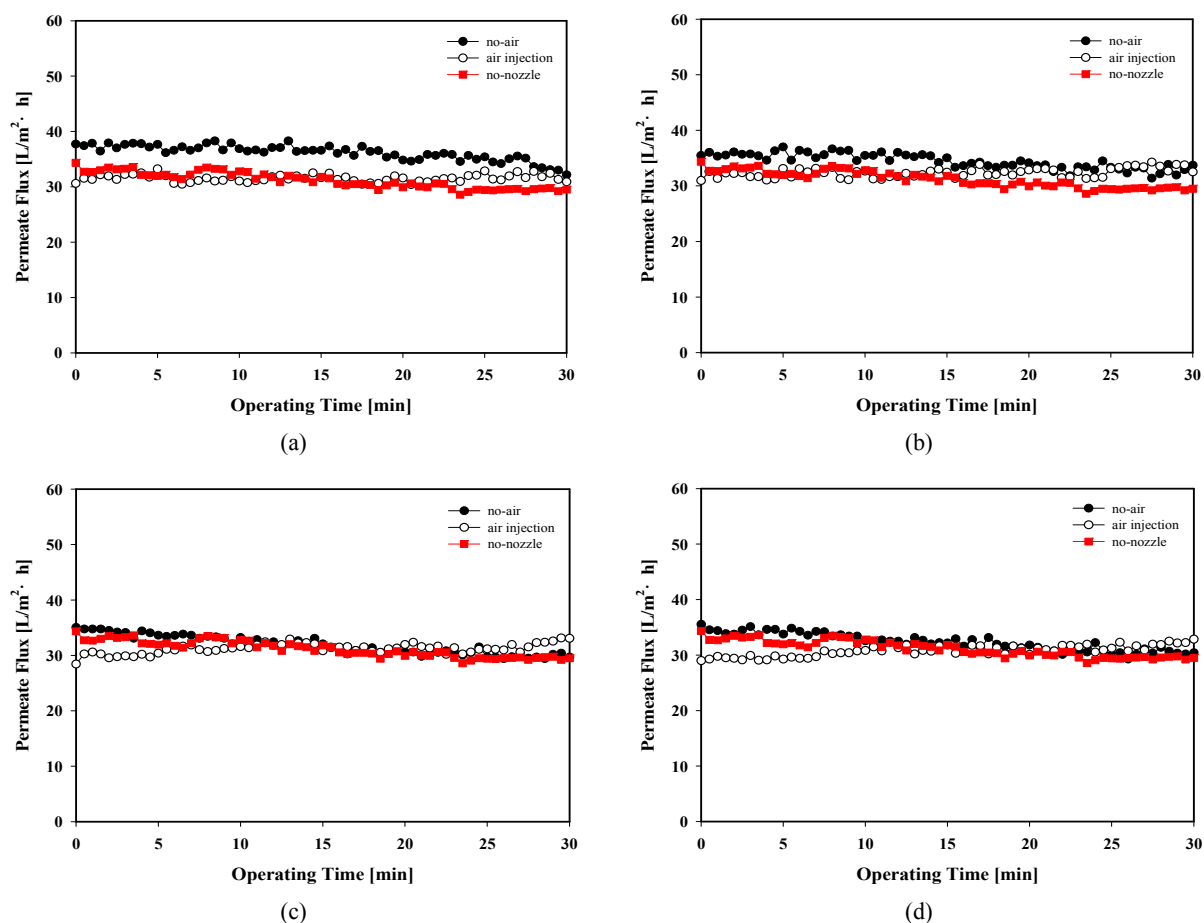
Fig. 6. Permeate fluxes as a function of gas flowrate for 0.1 wt% yeast solution with 0.1 μm tubular membrane at 0.8 bar, 20°C and (a) 1 L/min, (b) 2 L/min, (c) 3 L/min and (d) 4 L/min.

air injection was reduced the yeast layer on the membrane surface formed during the previous experiment without air injection.

The normalized permeate fluxes and flux enhancement for the yeast solution concentrations are summarized in Table 2. The flux enhancements for air in-jection relative to no-air or no-nozzle at 0.01 wt% yeast solution were 14 and 10%, respectively. The flux enhancement for air injection/no-nozzle was less than that for air injection/no-air at 0.01 wt% yeast solution. However, the flux enhancement for air injection/ no-nozzle was greater than that for air injection/no-air

Table 4. Normalized Permeate Fluxes (J/J_0) and Flux Enhancement for Yeast Solution for 0.1 μm Tubular Membrane at Various Liquid Flowrate

	Liquid flowrate	0.5 L/min	0.7 L/min	1.0 L/min	1.2 L/min
J/J_0	no-air	0.84	0.91	0.87	0.90
	air injection	1.06	1.09	1.02	1.11
	no-nozzle	0.93	0.86	0.89	0.89
Flux enhancement, %	air injection/no-air	26	20	17	23
	air injection/no-nozzle	14	27	15	25
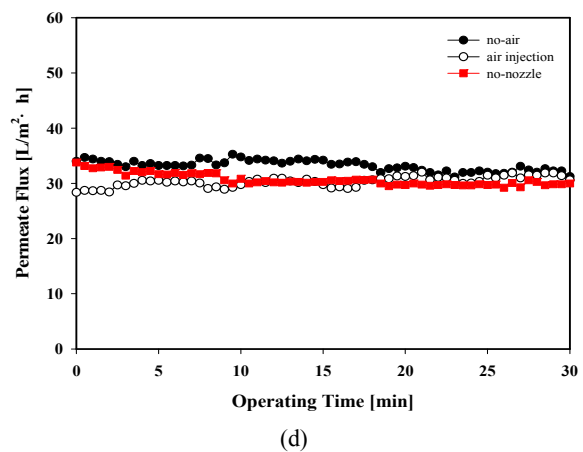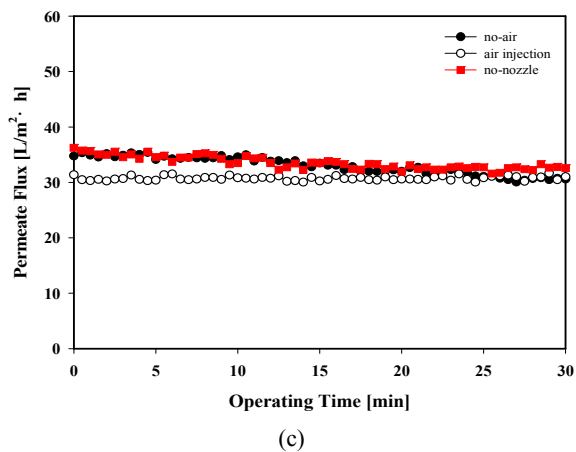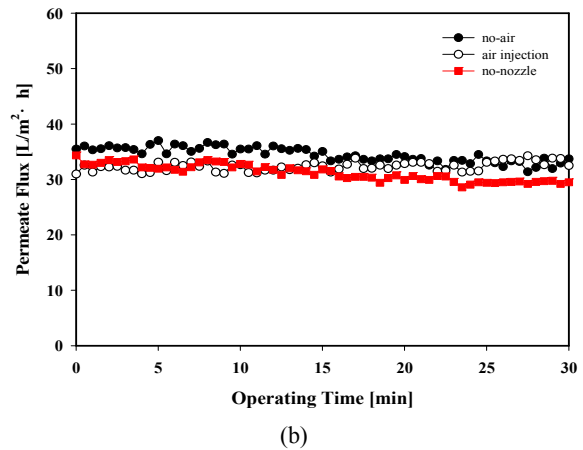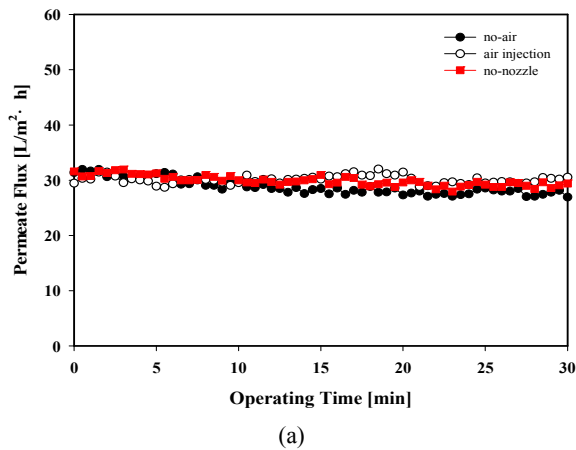
Fig. 7. Permeate fluxes as a function of liquid flowrate for 0.1 wt% yeast solution with 0.1 μm tubular membrane at 0.8 bar, 20°C and (a) 0.5 L/min, (b) 0.7 L/min, (c) 1.0 L/min and (d) 1.2 L/min.

above 0.05 wt% yeast solution.

3.3. Gas and liquid flowrates effect on flux enhancement

The major complexity in two-phase flows results from the growth and collapse of the gas-liquid inter-faces that can give to various flow regimes. Two-phase flows due to air sparging through the injection nozzle could change from the stratified-smooth to the inter-mittent: elongated bubble, slug and churn flow within the box as shown in Fig. 4. The moving elongated bubbles generate secondary flows behind the initial

bubble that assist in breaking up cake layer and subsequently promote local mixing near the membrane surface. The moving slugs result in pulsing pressure in the liquid around it, causing instability and disturbance near the membrane surface[20].

The permeate flux of 0.1 wt% yeast solution with respect to the gas flowrate are as shown in Fig. 6. The other operating condition for the variable of gas flowrate was fixed at 0.8 bar and 0.7 L/min liquid flowrate. The permeate fluxes for no-air and no-nozzle cases continuously decreased from 37.7 to 32.1 LMH and from 34.3 to 29.4 LMH at 1 L/min gas flowrate, respectively as shown in Fig. 6(a). The permeate flux for air injection case was maintained at about 31.5 LMH without a significant change. Two-phase flow patterns of 1 L/min gas flowates with 0.7 L/min liquid flowrate started from the only liquid flow at the beginning, and developed to the stratified-smooth flow regime. This developed two-phase flows might not be enough to defoul the yeast sufficiently on the membrane surface to recover the permeate flux. The permeate fluxes for no-air and no-nozzle cases continuously decreased from 36 to 32.9 LMH and from 34 to 29.4 LMH at 2 L/min gas flowrate, respectively as shown in Fig. 6(b). However, the permeate fluxes for air injection case increased from 30.9 to 33.7 LMH. As the gas flowrate increased to 2 L/min, two-phase flow patterns could be developed from liquid to the border of the stratified-smooth and the intermittent: elongated bubble, slug and churn flow condition which was strengthened defouling. As the gas flowrate increased further 3 or 4 L/min, the permeate fluxes for air injection increased from 28.4 and 28.9 LMH to 33 and 32.8 LMH, respectively. Above 3 L/min gas flowrate, the elongated bubble, slug and churn flow might be fully developed at the second half of the module, and induced more defouling on the membrane surface. The normalized permeate fluxes and flux enhancement for the gas flowrates are summarized in Table 3. Flux enhancements of air injection relative to no-air or no-nozzle case increased as the two-phase flow was changed from the stratified-smooth to elongated bubble,

slug and churn flow regime.

The permeate flux of 0.1 wt% yeast solution with respect to the liquid flowrate are as shown in Fig. 7. The other operating condition was fixed at 0.8 bar and 2.0 L/min gas flowrate. Two-phase flows due to air sparging through the injection nozzle could change the flow region from the stratified-smooth to the intermittent: elongated bubble, slug and churn flow within the box vertically as shown in Fig. 4. The permeate fluxes for no-air and no-nozzle cases continuously decreased from 31.9 to 26.9 LMH and from 31.6 to 29.4 LMH at liquid flowrate of 0.5 L/min, respectively as shown in Fig. 7(a). The normalized permeate fluxes and flux enhancement for the liquid flowrates are summarized in Table 4. Flux enhancements of air injection relative to no-air and no-nozzle case were more than 17 and 14%, respectively.

4. Conclusions

In this study, the tubular membrane module was self-designed with inserting the air injection nozzle tube in the module. The permeate fluxes were measured experimentally in order to evaluate the flux enhancement due to air injection, and the results are summarized as follows:

- The permeate fluxes of no-air injection with the nozzle tube were greater than those of the empty tubular module(no-nozzle). At lower operating pressure, the effect of flow eddies or mixing due to hindrance of the injection tube was greater than that of air injection. As operating pressure increased, air injection on the membrane surface through the air nozzle was observed relatively effective.

- The permeate flux enhancement for air injection was relatively greater than 10% below 0.05 wt% yeast solution. However, the flux enhancement increased at least 20% above 0.1 wt% yeast solution.

- Two-phase flows due to air sparging through the injection nozzle were bounded between the stratified-smooth and intermittent : elongated bubble, slug

and churn flow within 1-4 L/min gas flowrate and 0.5-1.2 L/min liquid flowrate. Flux enhancements of air injection for the gas flowrate increased as the two-phase flow was changed from the stratified-smooth to elongated bubble, slug and churn flow regime.

In conclusion, the tubular membrane module system with air injection was so effective to reduce yeast fouling or recover the permeate flux.

Acknowledgement

Reference

1. J. W. Lee, S. H. Kook, S. J. Kim, and I. S. Kim, "Effect of intermittent pressure-assisted forward osmosis (I-PAFO) operation on colloidal membrane fouling and physical cleaning efficiency", *Membr. J.*, **26**, 273 (2016).

2. Y. K. Choi, H. C. Kim, and S. H. Noh, "Effects of fouling reduction by intermittent aeration in membrane bioreactors", *Membr. J.*, **25**, 276 (2015).

3. H. N. Jang, I. C. Kim, and Y. T. Lee, "Membrane permeation characteristics and fouling control through the coating of poly(vinyl alcohol) on PVDF membrane surface", *Membr. J.*, **24**, 276 (2014).

4. Z. F. Cui and K. I. T. Wright, "Gas-liquid two-phase cross-flow ultrafiltration of BSA and dextran solutions", *J. Membr. Sci.*, **90**, 183 (1994).

5. J. G. Choi and K. Y. Chung, "Permeation characteristics of the microfiltration tubular module using the discharged rod", *Membr. J.*, **19**, 285 (2009).

6. J. A. Howell, T. W. Field, and D. Wu, "Yeast cell microfiltration: flux enhancement in baffled and pulsatile flow systems", *J. Membr. Sci.*, **80**, 59 (1993).

7. I. G. Rácz, J. Groot, and R. Klaassen, "Mass transfer, fluid flow and membrane properties in flat and corrugated plate hyperfiltration modules", *Desalination*, **60**, 213 (1986).

8. M. Mercier and C. Fonade, "Air-sparged micro-

filtration of enzyme/yeast mixtures: determination of optimal conditions for enzyme recovery", *Desalination*, **148**, 171 (2002).

9. K. J. Hwang and C. E. Hsu, "Effect of gas-liquid pattern on air-sparged cross-flow microfiltration of yeast suspension", *Chem. Eng. J.*, **151**, 160 (2009).

10. T. M. Qaisrani and W. M. Samhalber, "Impact of gas bubbling and backflushing on fouling control and membrane cleaning", *Desalination*, **266**, 154 (2011).

11. T. W. Cheng, "Influence of inclination on gas-sparged cross-flow ultrafiltration through an inorganic tubular membrane", *J. Membr. Sci.*, **196**, 103 (2002).

12. Z. F. Cui, S. Chang, and A. G. Fane, "The use of gas bubbling to enhance membrane processes", *J. Membr. Sci.*, **221**, 1 (2003).

13. G. Qian, J. Zhou, J. Zhang, C. Chen, R. Jin, and W. Liu, "Microfiltration performance with two phase flow", *Sep. Purif. Techol.*, **98**, 165 (2012).

14. C. Cabassued, S. Laborie, and J. M. Laine, "How slug flow can improve ultrafiltration flux in organic hollow fibres", *J. Membr. Sci.*, **128**, 93 (1997).

15. P. Mikulášek, J. Cakl, P. Pospíšil, and P. Doleček, "The use of flux enhancement methods for high flux cross-flow membrane microfiltration system", *Chem. Biochem. Eng. Q.*, **14**, 117 (2000).

16. H. Fadaei, S. R. Tabaei, and R. Roostaazad, "Comparative assessment of the efficiencies of gas sparging and back-flushing to improve yeast microfiltration using tubular ceramic membrane", *Desalination*, **217**, 93 (2007).

17. M. J. Park and K. Y. Chung, "Permeation characteristics of the tubular membrane with continuous air cleaning system", *Membr. J*, **23**, 185 (2013).

18. C. Liu, S. Caothien, J. Hayes, T. Caothuy, T. Otoyo, and T. Ogawa, "Membrane chemical cleaning: from art to science", pp 1, Pall Co., USA (2010).

19. O. Bratland, "Pipe Flow 2: Multi-phase flow assurance", pp 11, e-book (2013).

20. V. Kippax, "MemPulse MBR system vs traditional MBR systems", http://www.thembrsite.com/features/mempulse-mbr-system-vs-traditional-mbr-systems, June 04 (2011).