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Enhancement of Permeate Flux by Gas Slugs for Crossflow Ultrafiltration in Tubular Membrane Module

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ABSTRACT

Flux enhancements by gas slugs for dextran T500 solutions ultrafiltrated in a ZrO_2/carbon tubular membrane module were measured and are discussed for various resistances of the concentration boundary layer. These resistances are functions of the liquid velocity, the transmembrane pressure, and the feed concentration in the liquid-phase ultrafiltration. When the boundary layer resistance is low, the flux enhancement by gas slugs is limited. For a liquid ultrafiltration system with a severe concentration polarization, or operated in conditions of low liquid velocity, high transmembrane pressure, and high feed concentration, flux enhancement by gas slugs is very significant if the gas velocity exceeds a certain threshold. This threshold gas velocity depends on the extent of the concentration polarization in the single liquid-phase ultrafiltration system. It is concluded that the same permeate flux obtained in single liquid-phase ultrafiltration with a higher crossflow velocity can also be achieved with a lower liquid velocity by introducing gas slugs of moderate velocity, and lead to reduced energy consumption.

Key Words. Gas slugs; Ultrafiltration; Flux enhancement; Tubular membrane

INTRODUCTION

Ultrafiltration is a pressure-driven membrane separation process. The working pressure, usually applied to the solution in the 0.7×10^5 to $7 \times$

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10^5 Pa range, provides the driving potential needed to force the solvent or the smaller solute to flow through the membrane while the larger solute is rejected by the membrane. The purpose of the operation of ultrafiltration of the crossflow type is usually to decrease the accumulation of the rejected solute on the membrane surface. Ultrafiltration of macromolecular solutions has become an increasingly important separation process, and its applications include the treatments of industrial effluents, oil emulsion wastewater, biological macromolecules, colloidal paint suspensions, medical therapeutics, etc. The rapid development of this process was made possible by the advent of anisotropic, high-flux membranes capable of distinguishing among molecular and colloidal species in the 0.001 to 10 μm size range.

The permeate flux of an ultrafiltration process is dominated by the phenomena of concentration polarization and membrane fouling. In the search for ways to decrease concentration polarization and membrane fouling in order to increase the permeate flux, many studies have dealt with such techniques as external centrifugal forces (1), electric forces (2), pulsatile flows (3), turbulence promoters (4, 5), combinations of pulsatile flows and turbulence promoters (6), secondary flows (7, 8), and gas–liquid two-phase flows (9–16).

The method of gas–liquid two-phase flow in membrane filtration is a simple and economic technique which enhances the permeate flux effectively. It has been confirmed that this method not only offers a stable and large permeate flux, but also saves energy for methane fermentation (9). The addition of air to the liquid stream increases turbulence on the membrane surface and suppresses the formation of the concentration boundary layer, leading to enhancement in the flux of the filtration process. Lee et al. (10) used air slugs to improve the filtration of bacterial cell suspensions. The permeate flux was improved up to 100% with a polysulfone ultrafiltration membrane (MWCO 300 kDa) and up to 30% with a 0.2-μm PVDF microfiltration membrane, respectively.

Cui and Wright (11) investigated the effect of air sparging on ultrafiltrating macromolecular solutions in a tubular membrane (MWCO 100 kDa) which was mounted vertically or horizontally. The maximum flux enhancement was 60% for undyed 162 kDa dextran, 113% for 162 kDa dyed dextran, and 91% for 69 kDa BSA solutions. It has been shown in the above cited work that flux enhancement increased with a growth in transmembrane pressure, but hardly changed with variations in liquid flow rates and feed concentrations; the permeate flux for the upward flow in the vertically mounted membrane was 10–20% higher than that in the horizontally installed membrane. Recently, Cui and Wright (12) obtained experimental flux data for gas–liquid two-phase crossflow ultrafiltration in the downward flow condition with a 50-kDa tubular membrane module, and showed that the flux increment was up to 320% for a 260 kDa dextran solution compared to the single liquid-phase ultrafiltration. They also concluded that flux enhancement is more significant for the liquid phase in laminar flow than in turbulent flow. Bellara et al. (13) employed a pilot-plant scale hollow-fiber module, and they investigated the use of gas–liquid two-phase crossflow to overcome concentration polarization in the ultrafiltration of macromolecular solutions. Their work showed that the flux enhancements were 20–50% for dextran and 10–60% for albumin, and the sieving coefficient of albumin was considerably reduced when the gas-sparging technique was used.

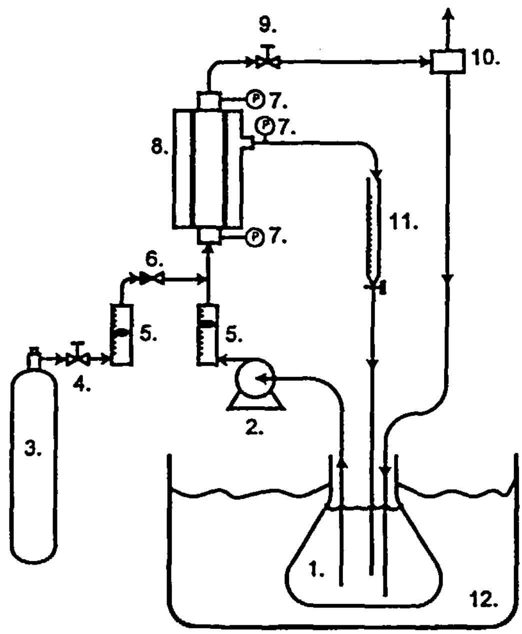
The flow pattern of gas–liquid two-phase flow is an important parameter for determining the performance of an air-sparging ultrafiltration system. Mercier et al. (14) consider the slug-flow pattern to be the most apropriate regime for increasing the efficiency of a gas–liquid two-phase filtration system. How the slug flow enhances the ultrafiltration flux in hollow fibers was studied by Cabassud et al. (15). Their experimental results showed that injecting the air leads to an increase of permeate flux by up to 110% for clay suspensions. Mercier et al. (16) investigated flux enhancement by gas slugs in the ultrafiltration tubular membrane, and the experimental results showed a 200% increase of flux for two kinds of suspension (bentonite and yeast).

This paper investigates the influence of gas slugs on flux enhancement for the upward cocurrent crossflow ultrafiltration of macromolecular solutions in a tubular membrane module with different liquid flow patterns (laminar and turbulent flow). The extent of concentration polarization phenomenon in a single-phase ultrafiltration system will be expressed as the resistance of the concentration polarization layer which is evaluated based on the boundary layer resistance model, and the flux enhancement is examined and discussed for various transmembrane pressures, liquid velocities, and feed concentrations.

EXPERIMENT

The experimental apparatus used in this work is shown in Fig. 1. This gas–liquid two-phase ultrafiltration system is operated in a tubular membrane module with the upward cocurrent flow. The membrane medium used was a 15-kDa MWCO tubular ceramic membrane (M2 type, Techsep, France) of 40.0 cm length, 6.0 mm internal diameter, and 75.4 cm^2 effective membrane area. The tested solute was dextran T500 (Pharmacia Co., Sweden) which was more than 99% retained by the membrane used. The solvent was distilled water.

The feed solution was circulated by a high-pressure pump with a variable speed motor (L-07553-20, Cole-Parmer Co.); the liquid flow rate was observed by a flowmeter (IR-OPFLOW 502-111, Headland Co.). The compressed air supply was directed to the liquid stream with the rate of gas



1. feed tank
2. pump
3. compressed air
4. air flow rate adjustment valve
5. flow meter
6. one-way valve
7. pressure gauge
8. membrane module
9. pressure control valve
10. gas/liquid separator
11. collector
12. thermostat

FIG. 1 Flow diagram of the gas–liquid two-phase crossflow ultrafiltration apparatus.

addition monitored by a flowmeter (F150-AV1-B-125-30-SAP, Porter Co.). The feed pressure was controlled by using an adjustable valve at the outlet of the membrane module, and the gauge pressures at the tubeside inlet (p_i), outlet (p_o) and at the shellside (p_p) were measured with a pressure transmitter (Model 891.14.425, Wika Co.).

The ranges of the experimental conditions were as follows. The feed concentrations, c_i, were 4.0–12.0 g/L; the liquid superficial velocities, u_i, were 0.15–0.90 m/s; the gas superficial velocities, u_{gs}, were 0.01–0.30 m/s; and the feed transmembrane pressures, Δp_i, were 58.8–196.0 kPa. The feed solution temperature in all experiments was kept at 30°C by a thermostat. As the liquid superficial velocity (u_i) is less than 0.3 m/s, the liquid flow is laminar; and when $u_i \geq 0.5$ m/s, the liquid flow is turbulent (17). During each run, both the permeate and the retentate were recycled back to the feed tank.

The experimental procedure was as follows. A fresh membrane was used to measure the permeate flux of water, J_w, for determining the intrinsic resistance of the membrane. The steady-state permeate flux for liquid solution ultrafiltration was measured first. Then the gas slugs, with a specified velocity, were injected into the liquid stream for obtaining the steady-state two-phase permeate flux, J_v.

After each experiment, the membrane was cleaned by high circulation and backflushing with 10% NaOH and 10% HNO_3 aqueous solutions and pure water. The cleaning procedure was repeated until the original water flux had been restored.

RESULTS AND DISCUSSION

Two-Phase Flow Patterns

When gas–liquid mixtures flow upward in a vertical tube with a small diameter, the two-phase flow may distribute in a number of patterns, each characterizing the radial and/or axial distribution of liquid and gas. According to the classifications by Taitel et al. (18) and Barnea et al. (19), as shown in Fig. 2, the two-phase flow patterns in this experimental range were mostly located in the slug flow region.

Intrinsic Membrane Resistance

The intrinsic resistance of a membrane, R_m, may be determined from the experimental data of the permeate flux data for pure water coupled with use of the equation

$$J_w = \frac{1}{\eta_w} \frac{\Delta P}{R_m} \tag{1}$$

where $\Delta P = (p_i + p_o)/2 - p_p$, is the mean transmembrane pressure and η_w is the water viscosity ($= 0.89 \times 10^{-3}$ Pa·s). With the use of experimental data, it was found that a straight line of $1/J_w$ vs $\eta_w/\Delta P$ could be constructed by the least-squares method with its slope showing the intrinsic resistance. The measured value of R_m for the membrane used was about 5.13×10^{12} m^2/m^3.

Permeate Flux of Liquid Ultrafiltration

For liquid ultrafiltration or $u_{gs} = 0$, Fig. 3 represents the relationship between the transmembrane pressure and the permeate flux for various liquid

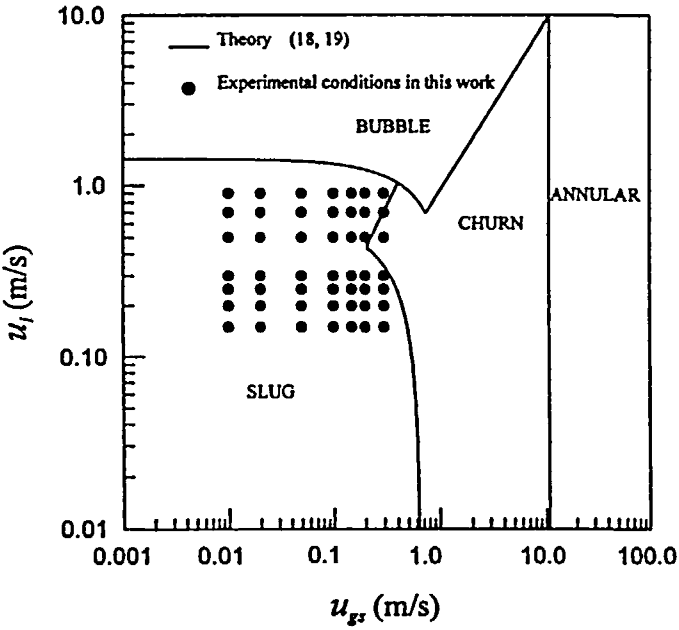


FIG. 2 Flow patterns map of gas–liquid two-phase flow.

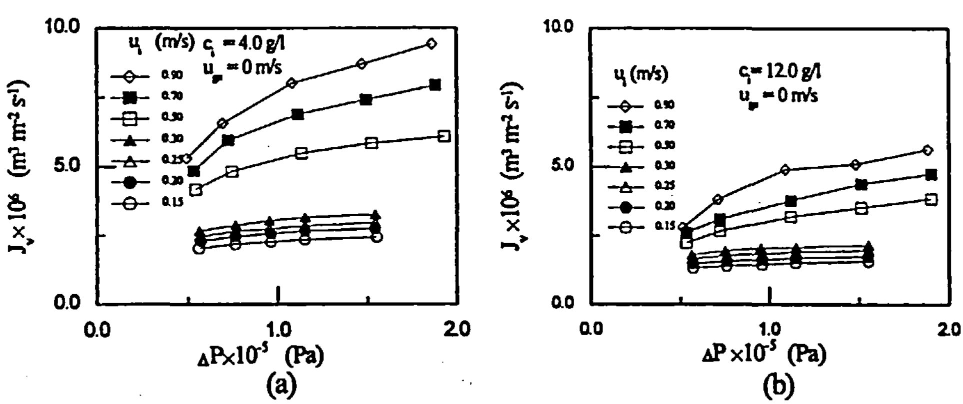


FIG. 3 Permeate flux of the single liquid phase ultrafiltration for various liquid velocities: (a) c_i = 4.0 g/L and (b) c_i = 12.0 g/L.

velocities. For the lower feed concentration solutions, the extent of concentration polarization is lower, so the permeate fluxes for $c_i = 4.0$ g/L (Fig. 3a) are higher than those for $c_i = 12.0$ g/L (Fig. 3b).

When liquid flow is laminar or $u_l < 0.3$ m/s, the permeate flux remained almost constant as the transmembrane pressure varied in the 50–150 kPa range for both feed concentrations, as shown in Fig. 3. Moreover, the increment in the permeate flux is small for such lower liquid velocities. This flux-limited behavior is due to severe concentration polarization on the membrane surface while the liquid velocity is too small to reduce it.

When liquid flow is turbulent or $u_l > 0.5$ m/s, the increment in the permeate flux is large as the transmembrane pressure increases. In these high turbulent conditions the rejected solutes cannot be deposited stably on the membrane surface and the permeate flux would not be limited to an asymptotic value, even though the transmembrane pressure reached 190 kPa in this experimental work.

Permeate Flux of Gas–Liquid Two-Phase Ultrafiltration

The effects of gas slugs on the permeate flux were measured. Plots of permeate flux vs transmembrane pressure for various gas velocities as c_i = 4.0 and 12.0 g/L are shown in Figs. 4 and 5, respectively. These results show that gas slugs enhance permeate flux in the ultrafiltration process. It is clear that increasing either liquid velocity or gas velocity will enhance permeate flux. However, the increase of liquid velocity will require more pump power than the increase of gas velocity. Moveover, the permeate flux obtained for $u_l = 0.15$ m/s and $u_{gs} = 0.02$ m/s is larger than that for $u_l = 0.30$ m/s without gas slugs. Similarly, the permeate flux achieved for $u_l = 0.5$ m/s and $u_{gs} = 0.10$ m/s is larger than that for $u_l = 0.9$ m/s without gas slugs when the operating transmembrane pressure is more than 1.13×10^5 Pa. Accordingly, the same permeate flux obtained with a higher liquid velocity but without gas slugs can also be achieved with a lower liquid velocity and moderate gas velocity with gas slugs, leading to reduced energy consumption.

In Figs. 4(a) and 4(b), where the liquid flow is laminar, the phenomenon of limited flux is found in both single liquid phase and gas–liquid two-phase ultrafiltration systems. This is due to the extraordinary phenomena of concentration polarization. This phenomenon is caused by a low crossflow velocity on the membrane surface. The permeate flux was enhanced slightly by the increasing transmembrane pressure as a small amount of gas slugs was introduced, and a high jump in the flux was observed as the gas velocity increased beyond a threshold. In these cases, the threshold gas velocity was about 0.10 m/s.

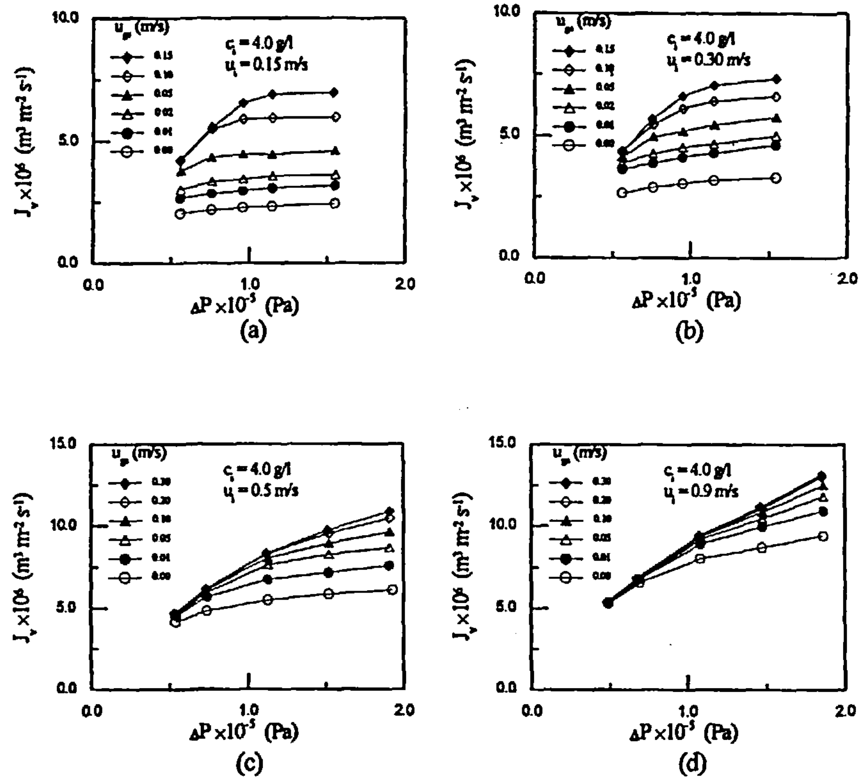


FIG. 4 Permeate flux of the gas–liquid two-phase ultrafiltration for various gas velocities as c_i = 4.0 g/L: (a) u_i = 0.15 m/s, (b) u_i = 0.30 m/s, (c) u_i = 0.50 m/s, and (d) u_i = 0.90 m/s.

In Figs. 4(d) and 5(d), where u_i = 0.90 m/s, no limited flux was observed because of high turbulence in this situation. The flux was enhanced notably even significantly when a small amount of gas slugs was introduced, especially in the condition of high transmembrane pressure, and the enhancement became asymptotic as the gas velocity was further increased. The work of Cui and Wright (12) showed that the effect of gas slugs was insignificant when the liquid flow was turbulent. This may be due to the fact that in their work the fluid velocity was high but the operating transmembrane pressure ($\Delta P = 1.0 \times 10^5$ Pa) was not high enough. It is seen in Figs. 4(d) and 5(d) that when the liquid velocity is high, the effect of gas slugs on permeation occurs only under high transmembrane pressure.

As shown in Figs. 4 and 5, though the influence of gas slugs on the flux behavior for c_i = 4.0 g/L and 12.0 g/L is similar, a difference between these two cases was found. At the liquid velocity of u_l = 0.15 m/s, the gas velocity required to achieve a large jump in flux enhancement is between 0.10 and 0.15 m/s for c_i = 12.0 g/L, as shown in Fig. 5(a), which is higher than the gas velocity (0.05–0.10 m/s, shown in Fig. 4a) for c_i = 4.0 g/L. A significant concentration polarization exists in the case of a high feed concentration and a low liquid velocity. Therefore, a higher gas velocity is required to disturb the concentration boundary layer and enhance the flux.

Flux Enhancements for Various Liquid Velocities

The enhancement of the permeate flux by gas slugs may be defined by a flux enhancement ratio, E, as

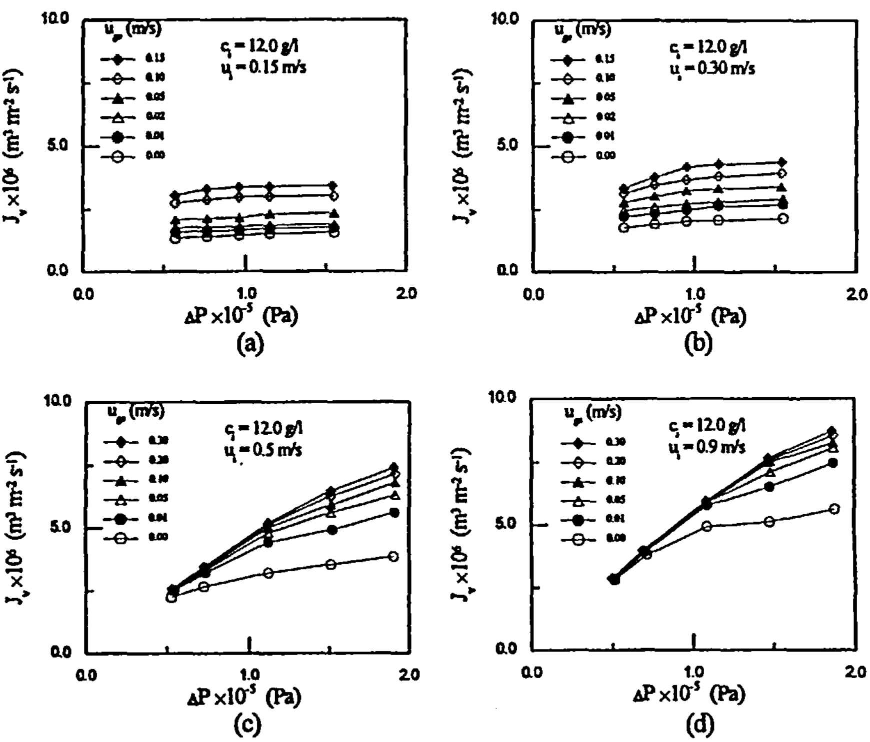


FIG. 5 Permeate flux of the gas–liquid two-phase ultrafiltration for various gas velocities as c_i = 12.0 g/L: (a) u_l = 0.15 m/s, (b) u_l = 0.30 m/s, (c) u_l = 0.50 m/s, and (d) u_l = 0.90 m/s.

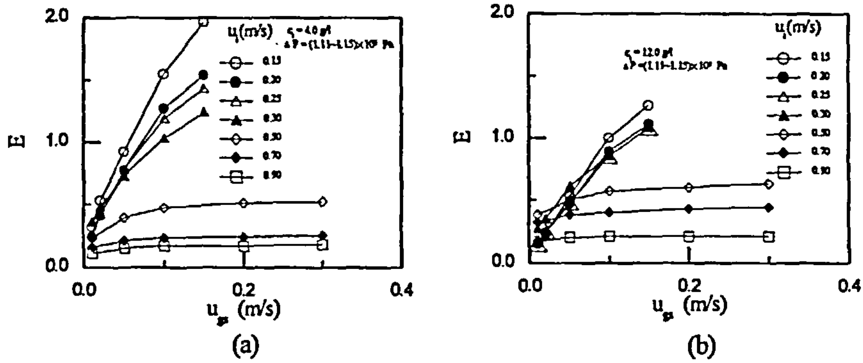


FIG. 6 Flux enhancements for various liquid velocities: (a) c_i = 4.0 g/L and (b) c_i = 12.0 g/L.

$$E = \frac{J_v - J_{v,0}}{J_{v,0}} \tag{2}$$

in which $J_{v,0}$ is the permeate flux without gas slugs. The highest enhancement ratio in this experimental work was 196%. Figure 6 plots the flux enhancement ratio vs gas velocity for various liquid velocities.

When the liquid flow is laminar (u_i < 0.3 m/s), the flux enhancement ratio is significant and increases as the gas velocity increases for both feed concentrations. For the lower feed concentration (see Fig. 6a), the increment in the flux enhancement ratio is more significant and increases as the liquid velocity decreases. But for the higher feed concentration (see Fig. 6b), the flux enhancement ratio responds only a little to a change in the liquid velocity. In this laminar liquid flow and under a high transmembrane pressure, a gel layer may be formed on the membrane surface. With a small drag force of the crossflow liquid, it is rather hard to remove the severe polarization layer on the membrane surface. It becomes even harder when the extent of concentration polarization increases. Therefore, the enhancement in flux due to gas slugs was both irregular and significant.

When the liquid flow is turbulent (u_i > 0.5 m/s), the flux enhancement ratio increases as the liquid velocity decreases. In the higher liquid flow rate operation, the accumulation of rejected solute as well as the concentration polarization on the membrane surface is relatively lower; therefore, flux enhancement by gas slugs is less effective. It becomes flat as the gas velocity increases further. Liquid velocities higher than 0.9 m/s were not treated in this work, but we believe that the flux enhancement obtainable by gas slugs in such a velocity region would be small.

Flux Enhancements for Various Transmembrane Pressures

Figure 7 plots the flux enhancement ratio vs gas velocity for various transmembrane pressures as c_i = 12.0 g/L. When u_i = 0.20 m/s and the liquid flow is laminar, the flux enhancement ratio increases as the gas velocity increases, but changes slightly with transmembrane pressure, as shown in Fig. 7(a). In Fig. 7(b) where u_i = 0.50 m/s and the liquid flow is turbulent, the flux enhancement ratio is smaller and turns out to be independent of a change in the gas velocity, especially for lower transmembrane pressures. Besides, the flux enhancement ratio increases with the transmembrane pressure. This result was also shown in Cui's work (11). Because of the significant concentration polarization under high transmembrane pressure, the increase in gas velocity still slightly improves the permeate flux for turbulent liquid flow when the operating transmembrane pressure is above 1.12×10^5 Pa, as shown in Fig. 7(b).

Flux Enhancements for Various Feed Concentrations

Figure 8 plots the flux enhancement ratio vs gas velocity for various feed concentrations. When the liquid ultrafiltration system is operated in the laminar flow region, as shown in Fig. 8(a), the flux enhancement ratio decreases with an increase in the feed concentration. For the conditions with low liquid velocity and high feed concentration, the introduction of a moderate amount of gas slugs is unable to disturb effectively the dense concentration polarization layer. Therefore, flux enhancement is smaller for the case with a higher

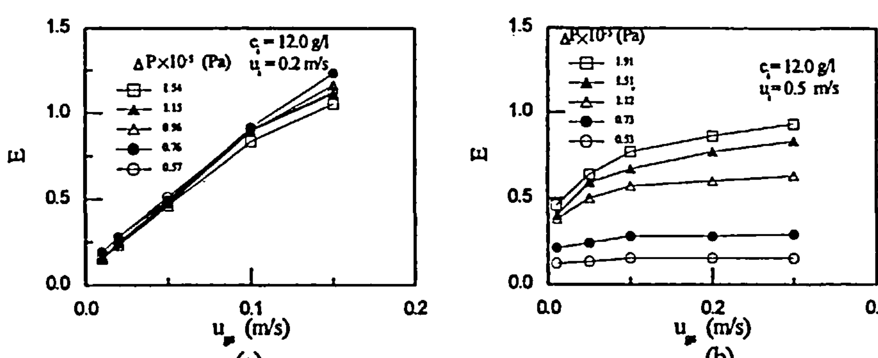


FIG. 7 Flux enhancements for various transmembrane pressures as $c_i \doteq 12.0$ g/L: (a) u_i = 0.20 m/s and (b) u_i = 0.50 m/s.

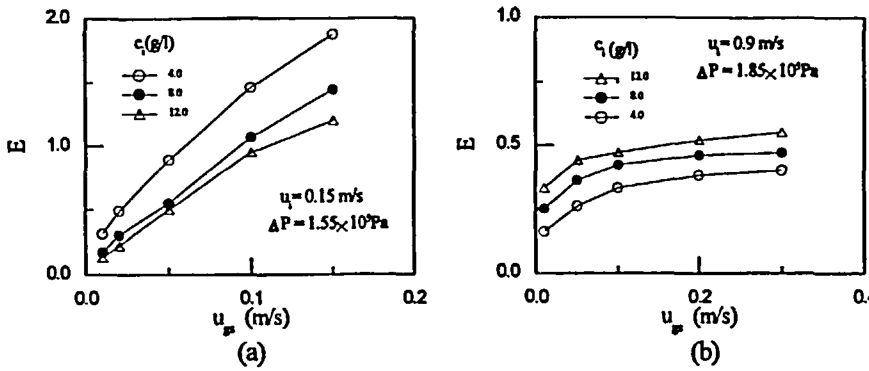


FIG. 8 Flux enhancements for various feed concentrations: (a) laminar flow region and (b) turbulent flow region.

feed concentration. But when the liquid ultrafiltration system is operated in the turbulent flow region, as shown in Fig. 8(b), the enhancement ratio increases with an increase in the feed concentration. This is because when the feed concentration is higher, the permeate flux is low in single liquid-phase ultrafiltration, so the flux enhancement ratio is larger compared to that with a lower feed concentration. It is also noted in Fig. 8(b) that the permeate flux is enhanced equally for various feed concentrations in this high liquid flow condition.

Flux Enhancements for Various Boundary Layer Resistances

As shown above, the effects of gas slugs on the flux enhancement ratio depend on the extent of concentration polarization in the single liquid-phase ultrafiltration system. The phenomenon of concentration polarization in the ultrafiltration process is a function of liquid velocity, the transmembrane pressure, and feed concentration. The resistance of the concentration polarization layer in a single liquid-phase ultrafiltration is defined by the boundary layer resistance model (20) as

$$J_{v,0} = \frac{1}{\eta_w} \frac{\Delta P}{R_m + R_{bl}} \tag{3}$$

in which R_{bl} is the boundary layer resistance. The values of R_{bl} in the experimental system can be evaluated from the experimental flux data of liquid

TABLE 1
Relation among Boundary Layer Resistances and Operating Conditions

c_i ($g \cdot L^{-1}$)	u_i ($m \cdot s^{-1}$)	$\Delta P \times 10^{-5}$ Pa	$J_{v,0} \times 10^6$ $m^3 \cdot m^{-2} \cdot s^{-1}$	$(R_m + R_{bl}) \times 10^{-12}$ $m^2 \cdot m^{-3}$	$R_{bl} \times 10^{-12}$ $m^2 \cdot m^{-3}$	Flow regime
12.0	0.9	1.09	4.91	24.91	19.77	Turbulent
8.0	0.9	1.87	6.69	31.20	26.07	Turbulent
12.0	0.5	1.12	3.19	39.18	34.05	Turbulent
12.0	0.5	1.51	3.52	47.82	42.68	Turbulent
12.0	0.5	1.90	3.84	55.48	50.35	Turbulent
4.0	0.2	1.54	2.73	63.16	58.03	Laminar

ultrafiltration and with the use of Eqs. (1) and (3). Some values of R_{bl} were calculated and are listed in Table 1.

Figure 9 plots the effect of gas velocity on the flux enhancement ratio for various boundary layer resistances. When R_{bl} is low, the flux enhancement ratio is small and the effect of gas slugs is limited with respect to different

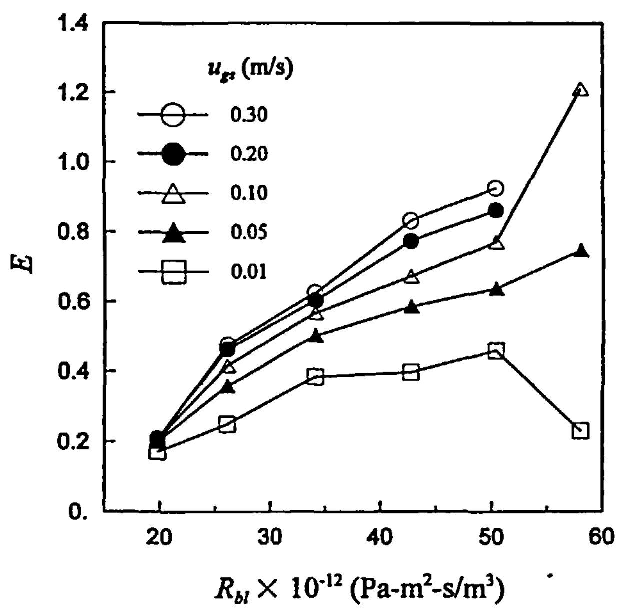


FIG. 9 Flux enhancements for various boundary layer resistances.

gas velocities. As R_{bl} increases, the flux enhancement ratio increases with an increase of gas velocity. Under this moderate concentration polarization condition, the gas slugs always enhance the permeate flux, and the benefits of gas slugs on flux enhancement are significant. For the case of severe concentration polarization (or R_{bl} is high), a low gas flow rate cannot effectively distrub the concentration polarization layer, so a gas velocity threshold is required to disturb the concentration polarization layer. Beyond this critical gas velocity, gas slugs can enhance the permeate flux significantly.

CONCLUSIONS

The effects of gas slugs on permeate flux were studied experimentally for the ultrafiltration of dextran T500 solutions in a crossflow tubular membrane module. Flux enhancement by gas slugs was measured and discussed for various resistances of the concentration boundary layer. These resistances are functions of liquid velocity, transmembrane pressure, and feed concentration. It was found that the same permeate flux in a single liquid-phase ultrafiltration with a higher crossflow velocity can also be achieved with a lower liquid velocity by introducing gas slugs of moderate velocity, which leads to reduced energy consumption.

When the boundary layer resistance (R_{bl}) is low, or when the operation is conducted under high liquid velocity, low transmembrane pressure, or low feed concentration, flux enhancement by gas slugs is limited with respect to various gas velocities. Under conditions of moderate concentration polarization, gas slugs enhance permeate flux effectively, and the flux enhancement ratio increases with an increase in gas velocity. For the case of severe concentration polarization, or for a liquid ultrafiltration system with low liquid velocity, high transmembrane pressure, and high feed concentration, flux enhancement by gas slugs is very significant if the gas velocity exceeds a threshold value. This gas velocity threshold depends on the extent of concentration polarization in a single liquid-phase ultrafiltration system. Determination of this critical gas velocity required to enhance the permeate flux in ultrafiltration systems with severe concentration polarization will be studied further.

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