특별강연 2

Enhancement of Pressure Driven Membrane Filtration by Natural Convection Instabilities

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자연대류 불안정성에 의한 압력추진 막여과 공정의 성능향상

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1. Introduction

In operation of the pressure driven membrane filtration systems, the loss of membrane performance, *i.e.* a drastic decline in flux with time is a major problem that should be solved for the successful application. The main reasons for the flux decline are concentration polarization and fouling. Especially, fouling which is a result of the irreversible deposition of solutes both at the membrane surface and in the membrane pores is mainly responsible for the loss of membrane performance.

To reduce or control concentration polarization and fouling, may methods have been suggested [1]. The most well known strategy is manipulating fluid flow in the membrane module such as increasing shear at the membrane surface by increasing cross-flow velocity or producing eddies by using turbulence promoters. One of the most effective approaches for manipulating fluid flow in the membrane module is to induce fluid instability near the membrane surface by using pulsate flow, and Taylor and Dean vortex flows [2,3].

An alternative method capable of inducing fluid instability in the membrane module is the use of natural convection. The natural convection is essentially generated in a stratified fluid in which higher-density layers overlay regions of lower-density. In the pressure driven membrane systems, the variation of solute concentration across the polarized layer means the existence of density variation so that the solution density at the membrane surface is higher than that in the bulk solution. By changing the gravitational orientation (inclined angle) of the membrane module, the density inversion in which higher-density solutions overlay lower-density solutions is obtained. This density inversion may lead to unstable fluid behavior and induce natural convection flow in the vicinity of the membrane surface. This concept is represented in Fig. 1. The occurrence of natural convection instability (NCI) flow in the membrane module may be effective to reduce concentration polarization and fouling because of its effects on promoting the back migration of solutes from the membrane surface to the bulk solution.

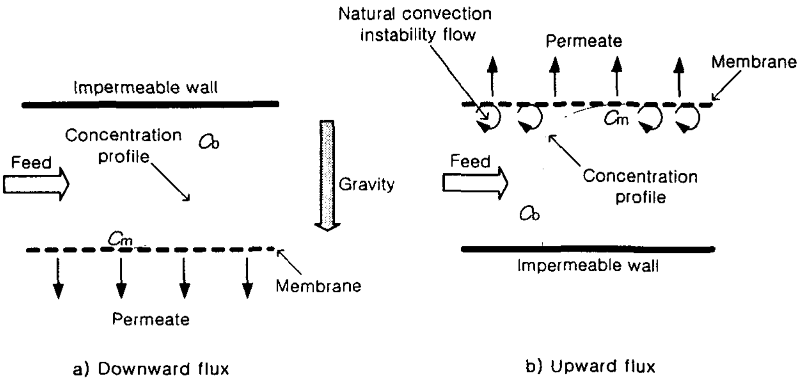


Fig. 1. Schematic representation of the occurance of natural convection instabilities in pressure driven membrane module, (a) downward flux (0°), (b) upward flux (180°).

This study is directed towards obtaining a better understanding of the efficacy of natural convection instability for the reduction of fouling and enhancement of performance in pressure driven membrane modules. Also it is directly relevant to our previous work [4] for the effects of NCI on ultrafiltration (UF) of relatively non-fouling dextran solute and significantly extends it. In this research, we studied systematically the effects of NCI on depolarization and defouling for a range of solutes that foul (protein) and form cakes (alumina and silica colloids, silica particle suspension) on the membrane thereby causing severe reduction in the flux. The UF performance of those solutes, according to the gravitational orientation of the membrane (from 0° to 180° inclined angle), was examined in an unstirred batch cell and a cross-flow module operated at both the constant-pressure and constant-flux mode. We evaluate the effects of NCI on membrane performance as the flux enhancement (E_i, in case of constant-pressure operation) and the reduction of transmembrane pressure (TMP) (R_i, in case of constant-flux operation). In addition, particular attentions have been paid to the influence of NCI on UF performance with change of the density and viscosity of protein solution and on the critical flux determination.

2. Experimental

2.1. Materials

Bovine serum albumin (BSA, molecular weight 69,000, Fraction V, from Sigma Co., USA) was used as the test protein and dissolved in salt free Milli-Q™ treated water ranged of concentrations from 0.2 to 5 kg/m^3. In some cases, the sucrose solutions that have been widely used as a gradient medium in density gradient ultracentrifugation were used as the dissolving solvent of BSA. Their use allows investigations of the NCI effects on UF performance with change of the density and viscosity of protein solution.

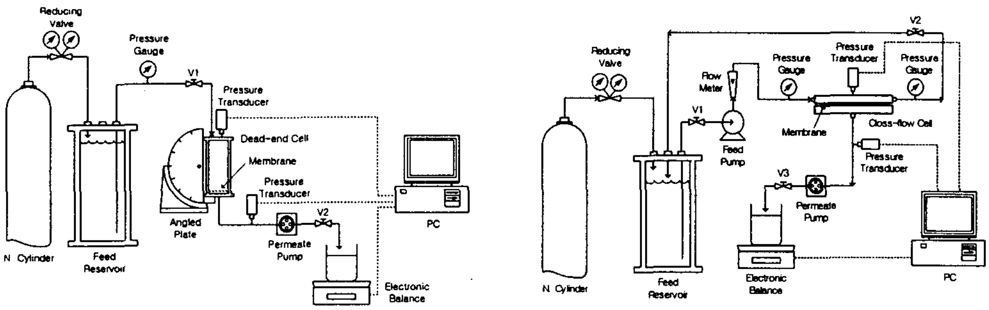
The commercial sols of alumina (Aluminasol-520, average particle diameter 15 nm) and silica (Snowtex PS-M, particle diameter 25 nm × particle length 125 nm) obtained from Nissan Chemical Co., Japan were used as the test sub-micron colloids. These stock sols (about 20 wt.% content) were diluted down to a concentration of 1 kg/m^3 with Milli-Q™ treated water. Also, silica particle (average particle diameter 4.6 ㎛, from Sambo Chemical Co., Korea) suspension dispersed in pure water was used as the test solute.

Polyethersulfone membrane (Biomax™ PBGC, MWCO = 10,000 Da, from Millipore Co., Australia) was used for the dead-end and cross-flow UF experiments to ensure almost complete rejection for the test solutes (BSA and sub-micron colloids) and complete permeation for the sucrose having low molecular weight.

2.2. Apparatus

A schematic diagrams of the dead-end and cross-flow membrane filtration experimental rig are shown in Fig. 2. Experiments were performed using a noncommercial unstirred batch cell (total capacity 130 ml, effective membrane area 15.2 cm^2) and cross-flow module (channel dimension: L 150 mm × H 3 mm × W 38 mm). The batch cell and cross-flow module fully charged with the test solution was placed on the angled plate for changing the gravitational orientation (from 0° to 180° inclined angle) of the membrane. The compressed nitrogen gas was applied on the feed reservoir to supply the test solution and operating pressure into the membrane cell. A personal computer was used to on-line acquire of the following experimental data: two pressure transducers one (CB1020, operating range 0~160 kPa, from Labom AG, Germany or PTX

500, operating range 0~700 kPa, from Druck Co., UK) at the roof of the membrane cell and the other (CB1020, operating range -50~100 kPa, from Labom AG, Germany) on the permeate line, and an electronic balance (BP610, from Sartorius AG, Germany) which measured flux by timed collection of the permeate. A permeate pump (Minipuls^{TM} 3 peristaltic pump, from Gilson Co., USA) was used to control the permeate flow rate when the constant-flux operating mode was specified. The pump speed was manually controlled.



(a) Dead-end rig (b) Cross-flow rig

Fig. 2. Diagrams of (a) the dead-end and (b) the cross-flow experimental rig.

2.3. Methods

Dead-end and cross-flow UF experiments under various gravitational orientation of the membrane cell (from 0° to 180° inclined angle) were performed in two operating modes: the constant-pressure and the constant-flux mode.

When the constant-pressure mode was required, the membrane cell was connected to a feed reservoir pressurized by nitrogen for adjusting the operating transmembrane pressure (TMP) without the permeate pump operation. In a typical constant-pressure run, the membrane permeability was first measured using pure water, and then the feed solution was fully charged into the membrane cell at the specified TMP (varied from 50 to 300 kPa) and gravitational cell orientation. Then the permeate flux as a dependent variable was recorded at 1- or 2-min intervals.

When the constant-flux mode was required, the permeate pump was used to control the flux (varied from 1.7 to 35 l/m^2 h). Prior to operate the permeate pump, the pressure in the membrane cell (upstream pressure) was adjusted at a fixed value (100 kPa for the protein solution and 50 kPa for the colloid) by connecting the membrane cell to a pressurized feed reservoir while the downstream valve (V2 or V3 in Fig. 2) was closed. This upstream pressure was continuously maintained for the entire duration of the constant-flux UF run. After the pressure of the permeate line reached the value of upstream pressure normally it takes about 30~40 minutes, the permeate pump was put into operation and the valve V2 or V3 was simultaneously opened and then TMP (difference in two pressure transducer readings) as a dependent variable was monitored at 2-min intervals. In some constant-flux UF runs, flux-stepping (incremental increase in flux at 30-min intervals) method was used to determine the critical flux at different orientation of the batch cell. All experiments were performed at room temperature (20°C).

A transmission electron microscope (Zeiss EM109, Germany) to view the cross-section of the membrane samples was used to show BSA fouling layers.

3. Results and discussion

We obtained the following important results:

(1) In the dead-end UF of BSA solution, changing the gravitational orientation (inclined angle) of the test cell induces NCI in the membrane module and higher inclined angle offers stronger NCI [see Figs. 3 and 4]. This induced NCI enhances back transport of the deposited solutes away from the membrane surface, therefore gives for the improvement of permeate flux (constant-pressure mode) [see Fig. 5] and the reduction of TMP (constant-flux mode) [see Fig. 6].

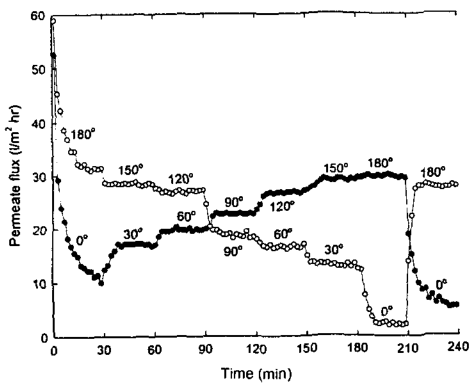


Fig. 3. Flux-time profiles for constant-pressure dead-end UF of BSA solution during successive change of the cell orientations [TMP = 200 kPa, feed concentration = 1 kg/m^3, (●); flux trends for starting at 0°, (○); flux trends for starting at 180°].

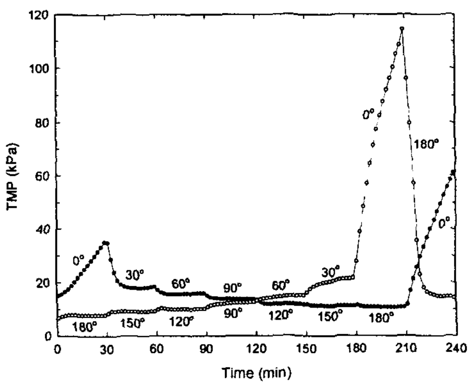


Fig. 4. TMP-time profiles for constant-flux dead-end UF of BSA solution during successive change of the cell orientations [flux = 11.4 l/m^2 h, upstream pressure = 100 kPa, feed concentration = 1 kg/m^3, (●); flux trends for starting at 0°, (○); flux trends for starting at 180°].

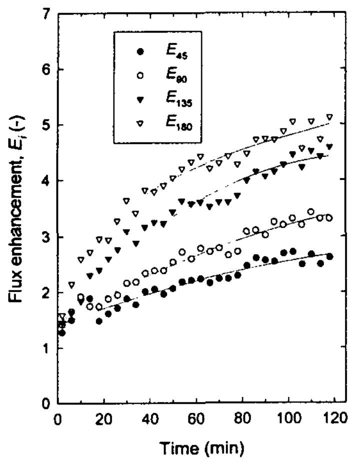


Fig. 5. Flux enhancement trends for constant-pressure dead-end UF of BSA solution at different cell orientations [TMP = 200 kPa, feed concentration = 1 kg/m^3].

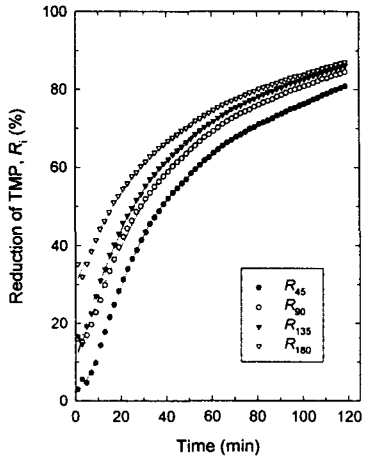


Fig. 6. Reduction of TMP trends for constant-flux dead-end UF of BSA solution at different cell orientations [flux = 10 l/m^2 h, upstream pressure = 100 kPa, feed concentration = 1 kg/m^3].

(2) In the dead-end UF of BSA solution, increasing the density of BSA solution brings decreasing the flux enhancement [see Fig. 7] and the reduction of TMP [see Fig. 8]. Such a phenomena appears to be caused by

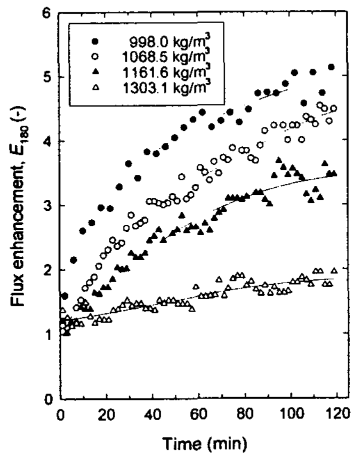


Fig. 7. Flux enhancement trends for constant-pressure dead-end UF at 180° angle with change of the density of BSA solution [TMP = 200 kPa, feed concentration = 1 kg/m^3].

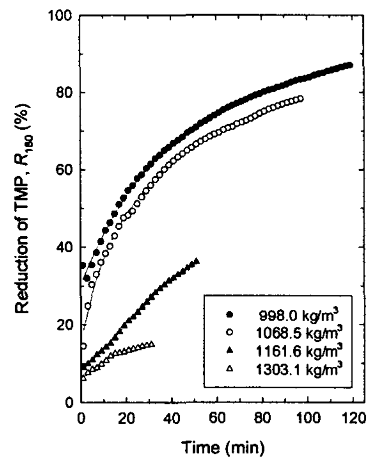


Fig. 8. Reduction of TMP trends for constant-flux dead-end UF at 180° angle with change of the density of BSA solution [flux = 10 l/m^2 h, upstream pressure = 100 kPa, feed concentration = 1 kg/m^3].

the gravitational force acting on the protein macromolecules comprising the polarized (or fouled) layer. Therefore, such an effect becomes less pronounced with the higher density of BSA solution, leading to a decrease in the difference between the density of BSA near the membrane surface and that of the bulk solution.

(3) In the dead-end UF and MF of colloidal suspensions, we obtained the following results. For alumina colloid (average size = 15 nm) [Fig. 9a], the effects of NCI occurrence on the flux are clear although the effects are less than that in case of BSA solution. However, for the silica colloid (average size = 75 nm) [Fig. 9b] and silica particle suspension (average size = 4.6 ㎛) [Fig. 9c], its effects are not appearing. These results clearly suggest that the size of colloidal particle affects the extent of NCI occurrence. More research for the effects of colloidal particle size on NCI occurrence is required.

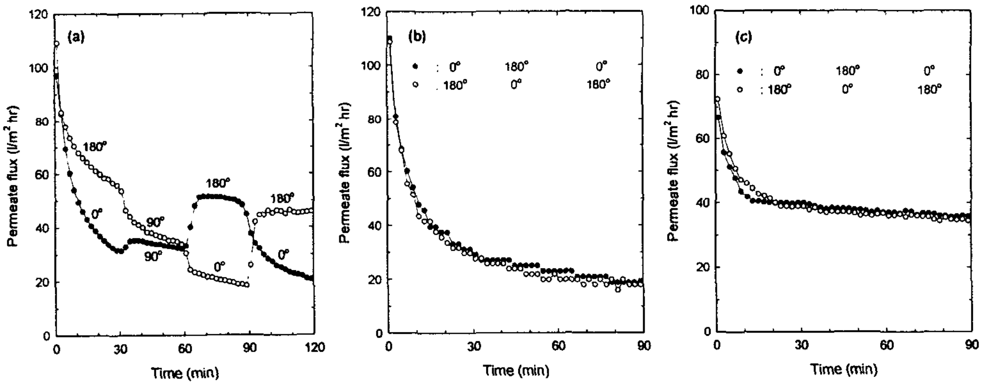


Fig. 9. Flux-time profiles for constant-pressure dead-end UF of (a) alumina colloid, (b) silica colloid and (c) silica particle suspension during successive change of the cell orientations [TMP = 100 kPa, feed concentration = 1 kg/m^3, (●); flux trends for starting at 0°, (○); flux trends for starting at 180°].

(4) In the dead-end UF of BSA and alumina colloid, the critical flux increases with increasing the inclined angle of the test cell [see Figs. 10 and 11]. These results also mean that NCI occurrence in the membrane module with the inclined angle enhances back transport of the deposited solutes away from the membrane surface, therefore gives for the increased critical flux.

(5) In the cross-flow UF of BSA solution, the flux improvement and the reduction of TMP by NCI occur when the cross-flow rate is below the value of around 10 l/h [see Figs. 12 and 13].

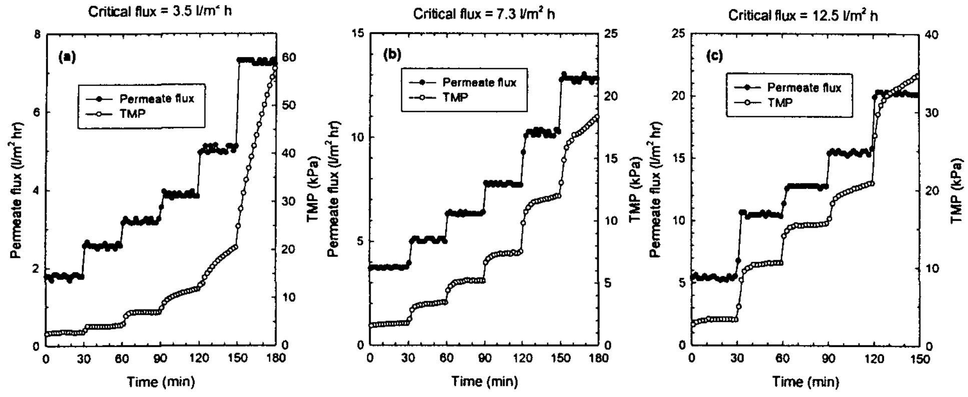


Fig. 10. Flux-stepping UF results of BSA solution for determination of critical flux at (a) 0°, (b) 90° and (c) 180° inclined angle [upstream pressure = 100 kPa, feed concentration = 1 kg/m^3].

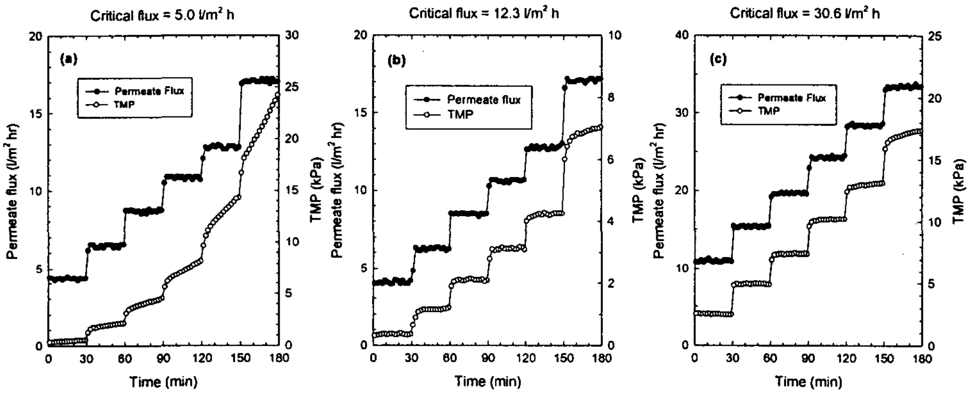


Fig. 11. Flux-stepping UF results of alumina colloid for determination of critical flux at (a) 0°, (b) 90° and (c) 180° inclined angle [upstream pressure = 50 kPa, feed concentration = 1 kg/m^3].

4. Conclusions

From these results, we conclude that being able to reduce fouling via simple techniques such as the induction of NCI in the pressure driven membrane modules offers significant economic benefits for the membrane processing operations. Savings may be realised via reduction in capital and/or operating costs. Reduced membrane fouling by NCI leads to enhanced flux and reduction in cleaning requirements for membrane modules. Enhanced performance results in increased module productivity or reduced membrane area and energy requirements for the fixed module productivity.

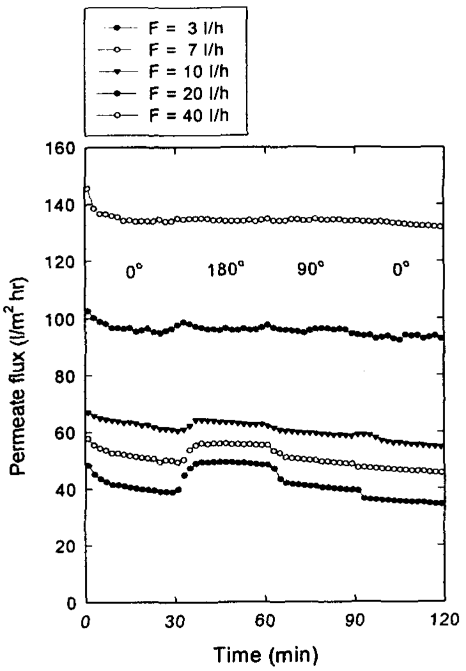


Fig. 12. Flux-time profiles for constant-pressure cross-flow UF of BSA solution during successive change of the cell orientations with change of the cross-flow rate [TMP = 200 kPa, feed concentration = 1 kg/m^3].

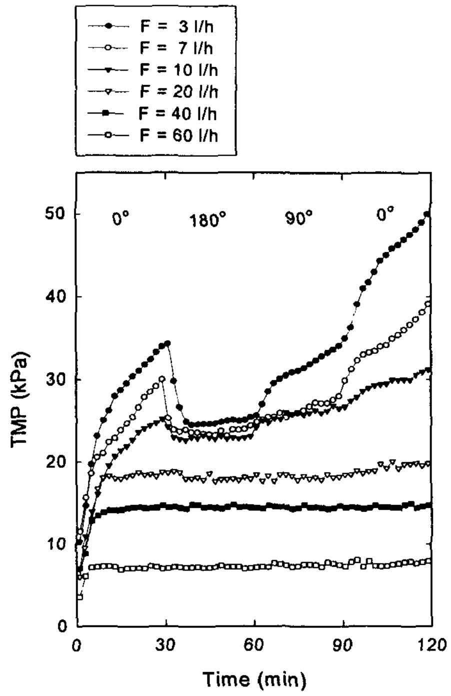


Fig. 13. TMP-time profiles for constant-flux cross-flow UF of BSA solution during successive change of the cell orientations with change of the cross-flow rate [flux = 17.5 l/m^2 h, upstream pressure = 110 kPa, feed concentration = 1 kg/m^3].

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