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Flux Enhancement in Cross-Flow Membrane Filtration by Flow Reversal: A Case Study on Ultrafiltration of BSA

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ABSTRACT

Fouling problems are perhaps the single most important reason for relatively slow acceptance of ultrafiltration in many areas of chemical and biological processing. To overcome the losses in permeate flux associated with concentration polarization and fouling, in cross-flow membrane filtration, we investigated the concept of flow reversal as a method to enhance membrane flux in ultrafiltration. Conceptually, flow reversal prevents the formation of stable hydrodynamic and concentration boundary layers at or near the membrane surface. Furthermore, periodic reversal of the flow direction of the feed stream at the membrane surface results in prevention and mitigation of membrane fouling. Consequently, these advantages are expected to enhance membrane flux significantly. BSA is a well-studied model solute in membrane filtration known for its

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fouling and concentration polarization capabilities. Laboratory-scale tests on a hollow-fiber ultrafiltration membrane module using bovine serum albumin (BSA) solution as feed show that under flow reversal conditions, the permeate flux is significantly enhanced when compared with the conventional unidirectional flow. The flux enhancement is dramatic (by an order of magnitude) with increased feed concentration and operating transmembrane pressure.

Key Words: Ultrafiltration; Flow reversal; Flux enhancement; BSA; Concentration polarization; Membrane fouling.

INTRODUCTION

In membrane-based separation, the terms “concentration polarization (CP)” and “membrane fouling” are always used to qualitatively and/or quantitatively to describe the flux decline. Specifically, in cross-flow membrane filtration (e.g., reverse osmosis, ultrafiltration, microfiltration, and nanofiltration), the loss of permeate flux with time of operation is inevitable. In many process plants, the productivity or the transmembrane flux in general is limited by the concentration polarization and fouling. The flux may be as low as 2 to 10% of that of pure solvent (water) flux in ultrafiltration membrane processes.[1]

The *concentration polarization* is viewed as the accumulation of dissolved solutes and macromolecules near or on the surface of the membrane due to convective and back-diffusive flow of solvent. As long as the particle or solute concentration at the membrane surface does not reach the maximum packing or gel concentration, the concentration polarization layer is mobile and does not offer a significant hydraulic resistance to permeate flow.[2] When the solute concentration reaches the gel concentration, a stagnant layer develops, which offers high resistance to permeate flow. The appreciable osmotic pressure in the polarized layer, due to the high local solute concentration, results in lowering the transmembrane pressure driving force. Manipulating the operating conditions can lessen the severity of concentration polarization.[3–5] The membrane *fouling* refers to the deposition of some feed components on the membrane surface and within the network of membrane pores.

In recent years, there has been renewed interest in understanding the underlying factors that limit the performance of cross-flow membrane processes and in finding a solution to the flux decline phenomena due to concentration polarization and membrane fouling. Surface modification or feed pretreatment has little effect on membrane flux due to secondary or gel layer formation.[6,7] To alleviate the deleterious effect of concentration polarization and membrane fouling, flow modifications in cross-flow membrane filtration are being studied as one of the most promising methods of choice.

The major emphasis in the design and operation of cross-flow filtration is to reduce the effects of concentration polarization and membrane fouling. It is now believed that to increase membrane flux, it is necessary to increase back transfer of solids from the membrane surface to the bulk solution. These are essentially based on the hydrodynamics and transport properties of the feed solution.[8–10] Some of the popular schemes that have been practiced or are being considered for flux enhancement in cross-flow filtration are shown in Fig. 1.

To minimize concentration polarization in cross-flow UF membrane modules, the conventional practice is to use high velocities at the cost of high-pressure drop, as shown in Fig. 1(a). With a rapid drop in pressure, transmembrane flux also drops rapidly with time. The problem is complicated by the fact that high-inlet pressure use would result in fouling by compaction at the inlet section of the module. On the other hand, a low pressure at the outlet leaves the outlet section of the membrane module underutilized, as shown schematically by the performance curve in Fig. 1(a). To overcome these limitations, periodic reversals of permeate flow back into feed channel or hollow-fiber lumen, known as "lumen flush," is an option practiced in many UF and MF operations. In the periodic lumen flush operation, the permeate flow valve is shut off for a few seconds, which forces permeate back into the feed channel. This results in dislodging accumulated particles or macromolecules from the membrane surface. As shown in Fig. 1(b), the pressure in the permeate side is about the average of feed side pressure since the feed flow is not shut off in lumen flush operation. As a result, only a section of the membrane module near the outlet is benefited, where the pressure in the permeate side is higher than the feed side. Thus, the method may be useful in some cases with limited success.[11]

An improved version of lumen flush is the periodic backwash (PBW), which is conducted by pumping permeates at higher pressure across the membrane to the feed side. This results in lifting or dislodging deposited materials from the membrane surface. As shown in Fig. 1(c), PBW can provide higher flux but its effectiveness may decrease with time, especially if pore fouling is the main cause.[12] In addition, it is to be noted that both in lumen flush and PBW, a fraction of the permeate is always lost due to flushing.

One modification of PBW mode of operation is to use uniform transmembrane pressure (UTP) accompanied by cocurrent permeate flow

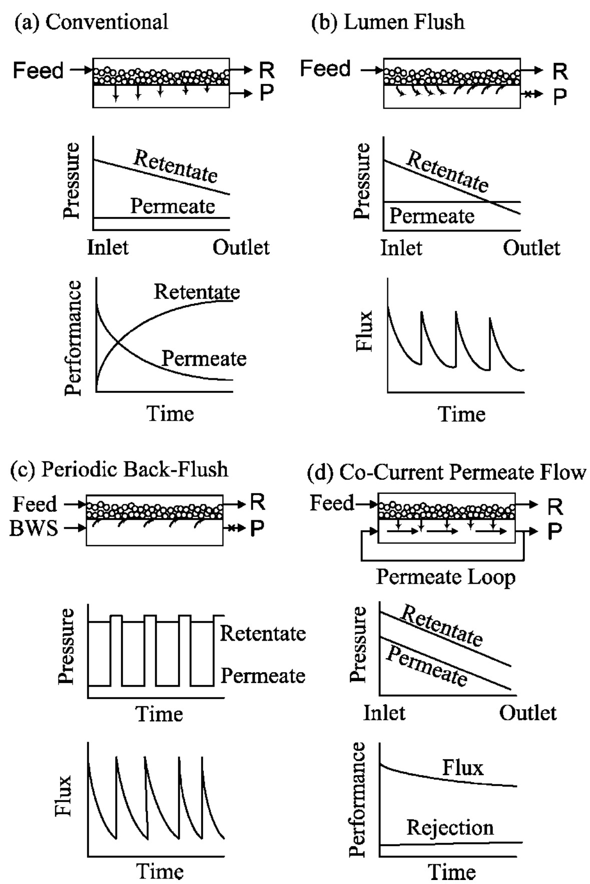
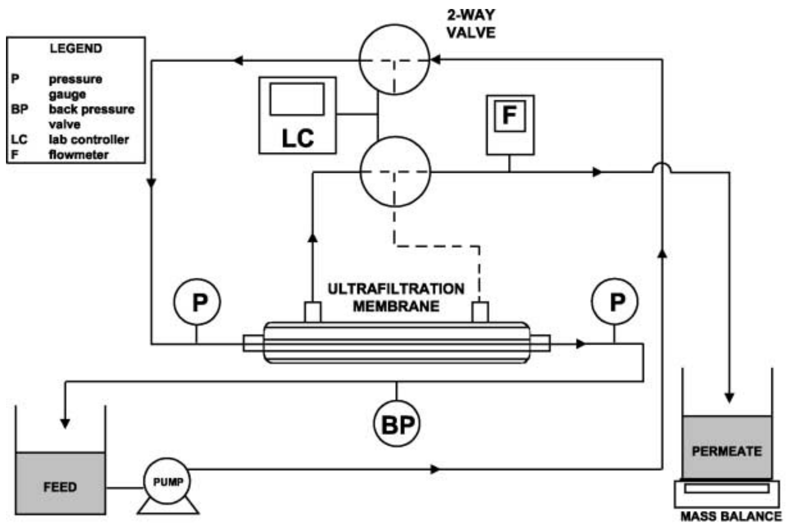
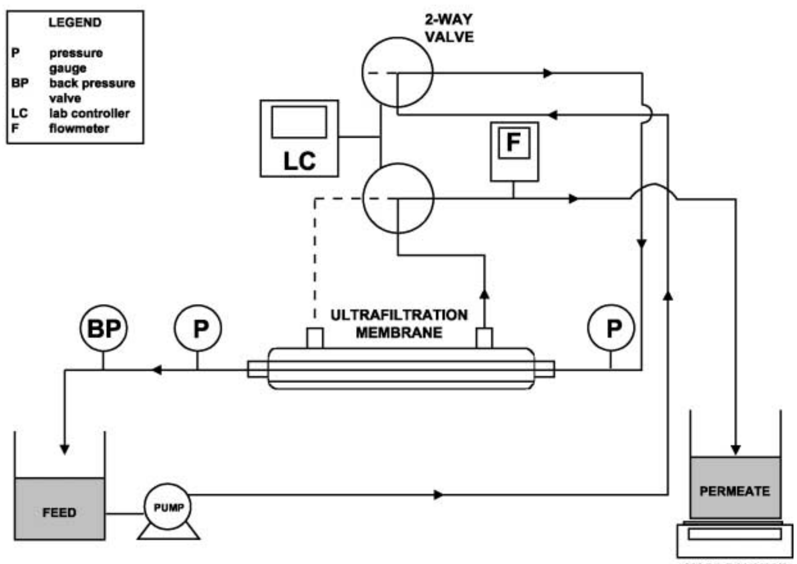


Figure 1. (a) Conventional, (b) lumen flush, (c) periodic back-wash, and (d) cocurrent permeate flow schemes in cross-flow membrane filtration, showing expected behavior of pressure profiles and time-dependent flux (P = permeate, R = retentate, and BWS = back-wash solvent).

(CPF). As shown in Fig. 1(d), this requires the simultaneous operation of a feed pumping loop and a permeate pumping loop to simulate a pseudo back-washing operation in a continuous manner, instead of periodic or intermittent backwash. With proper adjustment of two parallel flows of the feed and permeate, it is possible to maintain uniform transmembrane pressure, as shown schematically in Fig. 2(d). The UTP/CPF has been credited for enhanced flux in crossflow UF and MF operations.[13]



(a) Forward Feed Flow



(b) Reverse Feed Flow

Figure 2. Schematic of the experimental setup.

From this brief review, it is clear that various innovative methods have been proposed to overcome the limitations of concentration polarization and fouling. These have been partially successful, and in many situations, modifications were found to be difficult from engineering and economic considerations. To overcome the problems associated with concentration polarization and fouling, we investigated the concept of flow reversal as a method to enhance membrane flux in ultrafiltration.[14,15] Conceptually, flow reversal prevents the formation of stable hydrodynamic and concentration boundary layers at or near the membrane surface. Furthermore, periodic reversal of the flow direction of the feed stream at the membrane surface results in prevention and mitigation of membrane fouling. Consequently, these advantages are expected to enhance membrane flux significantly. The objective of this article is to report on some results of our ongoing work on flow reversal as an innovative method to enhance membrane flux by combating concentration polarization and fouling.

MATERIALS AND METHODS

Cross-flow membrane filtration experiments were conducted in tubular UF membrane modules using bovine serum albumin (BSA) as feed solution. The BSA, a well-studied model solute in membrane filtration, is known for its potent fouling and concentration polarization capabilities. The BSA solutions were prepared by dissolving appropriate amounts of bovine albumin fraction V powder in distilled water. The pH of the feed solution was not adjusted by adding any buffers. The Sigma Diagnostics Procedure No. 631 was used to determine the concentration of the BSA solution. The polysulfone UF membrane modules were obtained from A/G Technology (Amersham Biosciences Corporation, Piscataway, NJ, USA). The membrane module has an effective length of 31.5 cm, and contains 13 fibers, each with an internal diameter of 1 mm. The polysulfone membrane was rated at a nominal molecular weight cut-off of 3000.

The experimental set-up is shown schematically in Fig. 2. The forward feed flow and the reverse feed flow schemes are shown here. The forward feed flow scheme [Fig. 2(a)] is the one that is commonly used in cross-flow membrane filtration operation. By using two 2-way valves with the aid of a lab controller, the feed flow and the permeate flow directions can be switched at predetermined time intervals. The details of the experimental methods and materials used are reported elsewhere.[15]

RESULTS AND DISCUSSION

Cross-flow membrane filtration experiments were performed in a polysulfone UF tubular membrane module with BSA as a feed solution. The feed concentration ranged from 0.01 wt% to 5 wt% and the operating transmembrane pressure ranged from 20 psia to 30 psia. Transmembrane permeate flux data was collected for both the unidirectional and flow reversal conditions. For comparison purposes, unidirectional flow is considered as base or reference case. Each experiment was conducted for about 130 minutes. To maintain membrane performance, the membrane modules were thoroughly cleaned after each use according to manufacturer's cleaning procedure. Pure water flux data was collected initially for a new membrane and after each cleaning to ensure comparability of the experimental data.

The variation of permeate flux with time with and without flow reversal at a transmembrane pressure of 25 psia for 1.0 wt% and 3.0 wt% BSA feed solutions are shown in Figs. 3 and 4, respectively. The data show that there is a noticeable

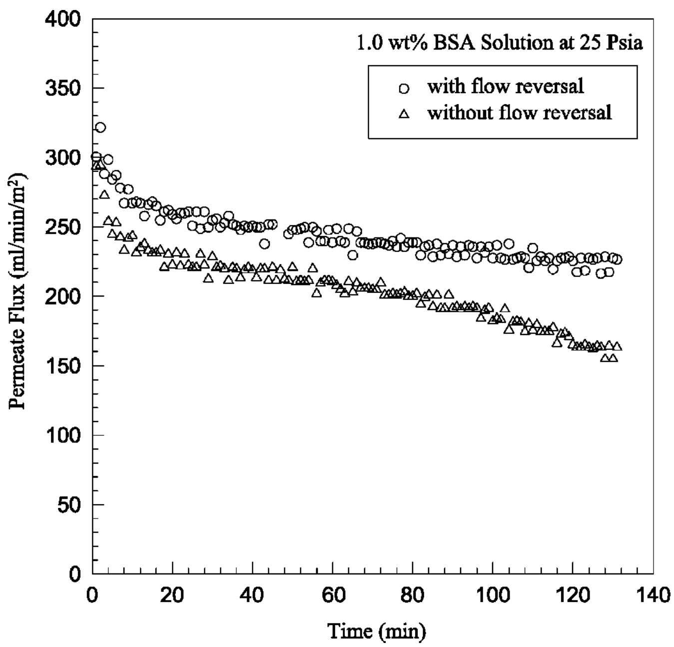


Figure 3. Comparison of permeate flux data for 1.0 wt% BSA solution at a transmembrane pressure of 25 psia.

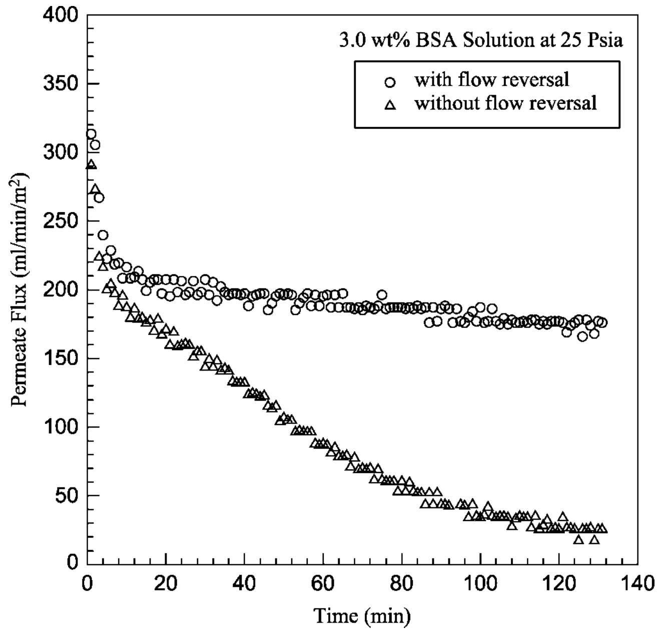


Figure 4. Comparison of permeate flux data for 3.0 wt% BSA solution at a transmembrane pressure of 25 psia.

gain in permeate flux with flow reversal. A comparison of the flux data in Figs. 3 and 4 show that the flux enhancement is significant at higher feed concentration. Without flow reversal, the flux declines very rapidly at higher feed concentration, as expected. However, with the flow reversal, the flux decline trend can be significantly slowed down with a net gain in permeate flux.

The flow reversal experiments were performed with a flow reversal time of 2 minutes, i.e., every 2 minutes, the direction of the feed and permeate flows were reversed using the computer-controlled valve manifolds. The flow switching time of 2 minutes was chosen because the flux decline in cross-flow filtration due to concentration polarization takes place in the first few minutes of operation. Therefore, the trick is to destabilize the concentration boundary by reversing the flow direction in a short interval of time. This helps to minimize the negative effect of concentration polarization on permeate flux. Furthermore, in absence of the stable concentration polarization layer or the gel layer, the membrane fouling is slowed down or further mitigated, with net gain in permeate flux over conventional cross-flow filtration.

Figures 5 and 6 show the flux data with time at a transmembrane pressure of 30 psia with 1.0 wt% and 3.0 wt% of BSA feed solutions, respectively. With increased solute concentration in the feed, one would expect rapid decline in permeate flux with time in conventional (base case) cross-flow filtration at higher transmembrane pressure. This is supported by the experimental flux data in Figs. 5 and 6 for the base case. If we compare the case of flow reversal with that of the base case, we observe that the gain in flux with flow reversal is phenomenal at higher transmembrane pressures. In fact, with 3.0% BSA feed solution at 30 psia operating transmembrane pressure, without flow reversal, the permeate flux drops to about 10 mL/min/m^2 in about 1 hour of UF operation (see Fig. 6). With flow reversal, the permeate flux can be maintained at about 200 mL/min/m^2 for a prolonged period of time.

Based on the experimental results presented, it can be seen that periodic reversal of flow of feed solution mitigates the effects of concentration polarization and membrane fouling that causes the initial rapid decline in permeate flux. The periodic reversal of the flow direction of the feed solution

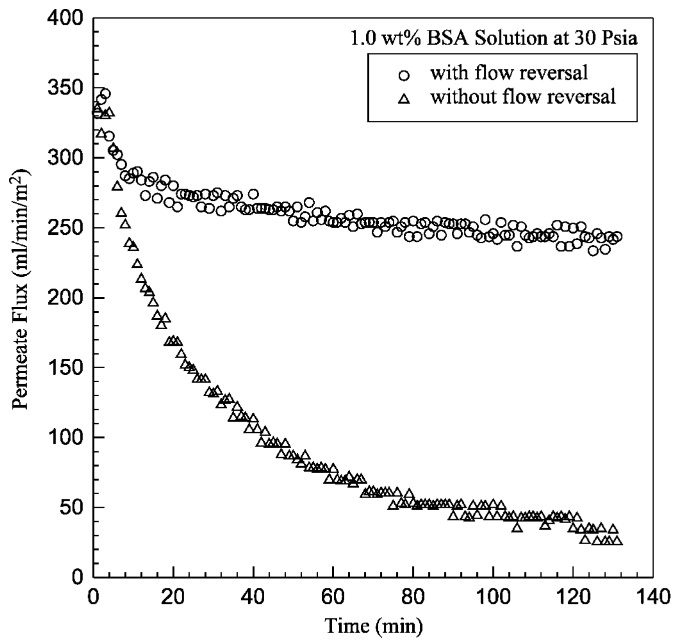


Figure 5. Comparison of permeate flux data for 1.0 wt% BSA solution at a transmembrane pressure of 30 psia.

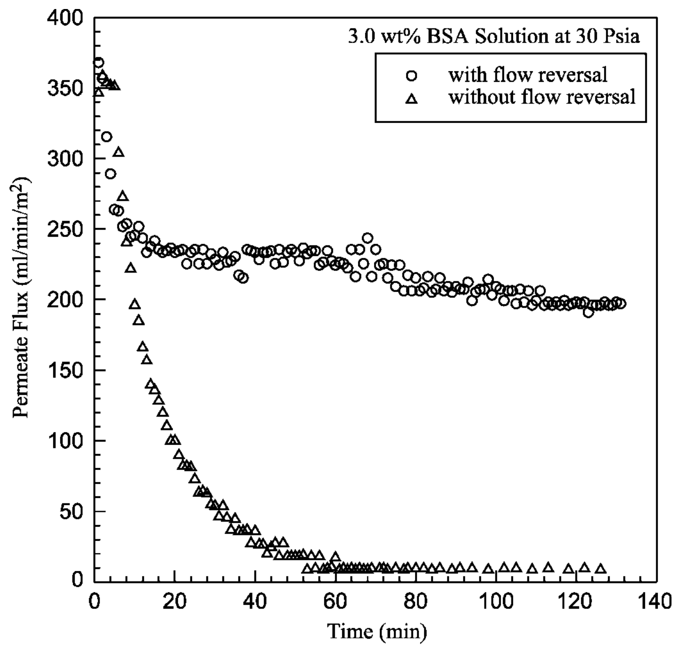


Figure 6. Comparison of permeate flux data for 3.0 wt% BSA solution at a transmembrane pressure of 30 psia.

at the surface of the membrane prevents the formation of stable hydrodynamic and concentration boundary layer. As the UF operation progresses over time and protein macromolecules are retained by the membrane, some adsorption is expected. However, the hydrodynamic instability by periodic flow reversal severely retards that adsorption. Hence, the collection of macromolecules at the membrane surface is significantly reduced. This results in enhanced permeate flux with the use of periodic flow reversal of the feed solution.

CONCLUSION

The concept of periodic reversal of feed flow in cross-flow UF operation for flux enhancement was investigated in a laboratory-scale tubular UF membrane module using BSA as feed solution. The results suggest that by flow reversal, significant enhancement of flux is possible and it can be used as an effective means to mitigate the deleterious effects of membrane fouling and concentration polarization.

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