Report Title:

Flux Enhancement in Crossflow Membrane Filtration: Fouling and It's Minimization by Flow Reversal

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DISCLAIMER

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. Further more, 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.

A crossflow membrane filtration unit was designed and built to test the concept of periodic flow reversal for flux enhancement. The essential elements of the system include a crossflow hollow fiber membrane module integrated with a two-way valve to direct the feed flow directions. The two-way valve is controlled by a controller-timer for periodic reversal of flow of feed stream. Another important feature of the system is that with changing feed flow direction, the permeate flow direction is also changed to maintain countercurrent feed and permeate flows for enhanced mass transfer driving force (concentration difference).

In our previous report, we reported our work on UF of BSA. In this report, we report our continuing application of Flow Reversal technique in clarification of apple juice containing pectin. The presence of pectin in apple juice makes the clarification process difficult and is believed to cause membrane fouling. Of all compounds found in apple juice, pectin is most often identified as the major hindrance to filtration performance. Laboratory-scale tests on a hollow-fiber ultrafiltration membrane module using pectin in apple juice as feed show that under flow reversal conditions, the permeate flux is significantly enhanced when compared with the conventional unidirectional flow.

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EXECUTIVE SUMMARY

To test the concept of period flow reversal of feed and permeate flows, crossflow membrane filtration unit was designed and built. The system was successfully tested for crossflow filtration of BSA in tubular UF membrane module. Currently, we are testing pectin in apple juice as new feed solution. Like BSA, we observed encouraging results with pectin in apple juice clarification utilizing flow reversal technique. These experimental tests 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.

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 crossflow membrane filtration (e.g. reverse osmosis, ultrafiltration, microfiltration and nano-filtration) 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 [Smolder and Boomgard, 1989].

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 pemeate flow [Redkar, et al., 1996]. 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 [Gekas and Hallstrom, 1987; Cheryan, 1998; Hargrove and Ilias, 1999]. 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 crossflow 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 [Brink and Romjin, 1990; Kim, et al., 1988]. To alleviate the deleterious effect of concentration polarization and membrane fouling, flow modifications in crossflow membrane filtration are being studied as one of the most promising methods of choice.

Recently, we investigated the use of flow reversal in cross-flow membrane UF as a means of increasing transmembrane flux by reducing the deleterious effects of concentration polarization and membrane fouling. BSA is a well-studied model solute in membrane filtration known for its fouling and concentration polarization capabilities [Hargrove and Ilias, 1999, Ilias, et al., 2001]. From limited UF experiments with BSA, we observed that flow reversal significantly improves the permeate flux and merits further research.

In this proposal, we address the membrane-fouling problem in crossflow ultrafiltration and microfiltration systems as follows:

- In membrane separation processes when dealing with multicomponent feed streams, no matter how good is the membrane properties and system design, flux decline due to fouling and concentration polarization is inevitable.
- Flux decline problem is a two step process: far field effects (hydrodynamic interactions) and near field effects (surface forces, chemical and electrokinetic interactions).

- Management and control of far field and near field effects can only give us adequate answer to the solution and control of membrane fouling problem.

Thus to develop a generic approach to reduce the membrane fouling, we consider a novel innovative technique to manipulate the far field hydrodynamics in such as way that solute convection-diffusion transport and particle migration to the membrane surface can never form a stable layer. If this can be achieved, a substantial increase in transmembrane flux would be possible. From our recent work on the feasibility of flow reversal as a technique to enhancing crossflow membrane fluxes, we found that the flow reversal has a great potential in combating flux reducing effects due to concentration polarization and fouling.

RESEARCH OJECTIVES

The objectives of this research are to:

1. Design and build a proto-type laboratory scale crossflow membrane filtration unit with periodic flow reversal option
2. Perform membrane filtration experiments with BSA, Dextran T-70 and FGD wastewater with and without periodic flow reversal option and compare performances.
3. Model the periodic flow reversal in crossflow filtration and validate the experimental observations with model predictions.

EXPERIMENTAL: MATERIALS & METHODS

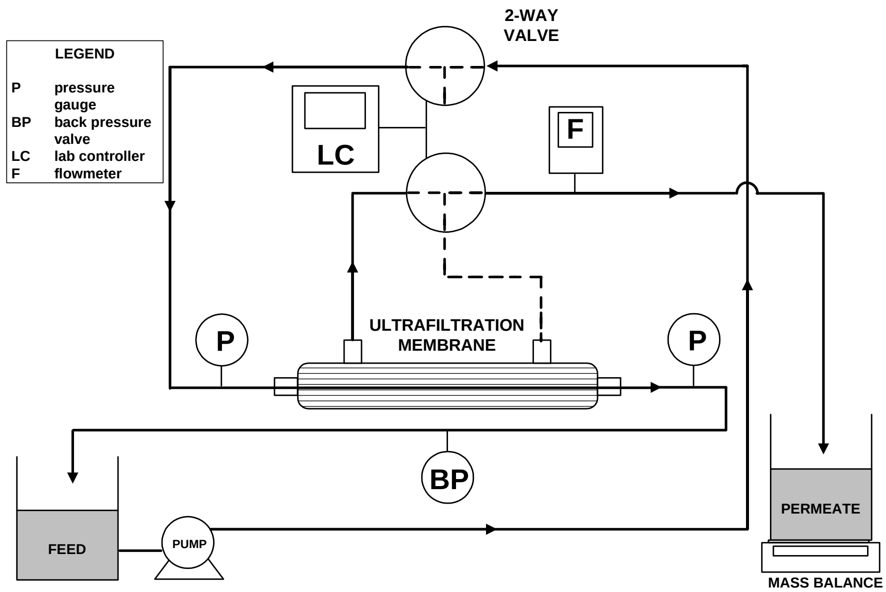
Cross flow membrane filtration experiments were conducted in tubular UF membrane modules using two feed solutions. In our previous report, we reported our work on bovine serum albumin (BSA) as feed solution. We are currently investigating the UF of apple juice with pectin using our flow reversal technology. Pectin is well known foulant in membrane filtration. In this report, we present some results on crossflow membrane filtration with and without periodic flow reversal with apple juice with pectin as feed solution.

The polysulfone UF membrane modules were obtained from A/G Technology. The membrane module has an effective length of 31.5 cm, and contains 13 fibers, each with an internal diameter of 1mm. The polysulfone membrane was rated at a nominal molecular weight cut-off of 3000.

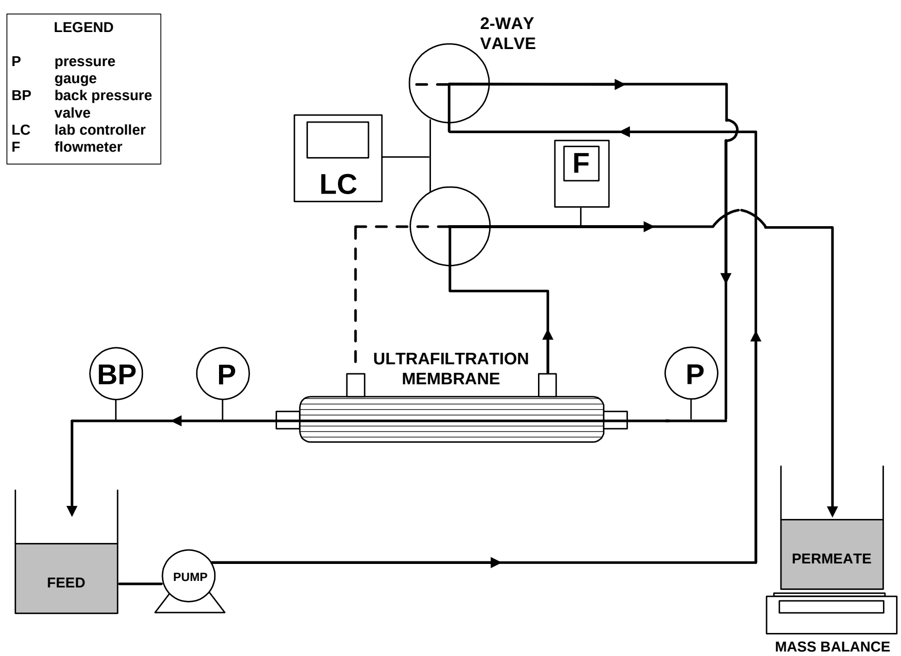
The experimental set-up is shown schematically in Figure 1. The forward feed flow and the reverse feed flow schemes are shown here. The forward feed flow scheme (Figure 1(a)) is the one that is commonly used in cross flow membrane filtration operation. The feed flow and the permeate flow directions were switched at predetermined time intervals using two 2-Way Valves, which was operated by a Lab Controller. The details of the experimental methods and materials used are reported elsewhere [Hargrove, 1998]. The experimental set-up is based on earlier patented work [Ilias, et al., 2002].

RESULTS AND DISCUSSIONS

During this reporting period, cross-flow membrane filtration experiments were performed in polysulfone UF tubular membrane module with apple juice as a feed solution. The pectin content in the apple juice ranged from 0.01 wt% to 0.05 wt% and the operating transmembrane pressure ranged from 20 Psia to 30 Psia [Pryor and Ilias, 2003]. Trans-membrane permeate flux data was collected for both the unidirectional and flow reversal conditions.



(a) Forward Feed Flow



(b) Reverse Feed Flow

Figure 1: Schematic of the experimental setup

For comparison purpose, unidirectional flow is considered as base or reference case. Each experiment was conducted for about two hours. 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 presence of pectin in apple juice makes the clarification process difficult and because of its fiber-like structure, which is believed to cause membrane fouling. Of all compounds found in apple juice, pectin is most often identified as the major hindrance to filtration performance. In Figure 2, the performance of UF of 0.05 wt% pectin in apple juice is shown. The UF operation was at transmembrane pressure difference, $\Delta P_{\text{TMP}} = 30 \text{ Psia}$, and feed flow Reynolds number, $N_{\text{Re}} = 2184.$ With conventional UF operation, permeate flux drops rapidly with time. The same operation using our flow reversal technology, with flow reversal time, $\tau_{rev} = 1 \text{ min}$, we find a marked improvement in transmembrane flux.

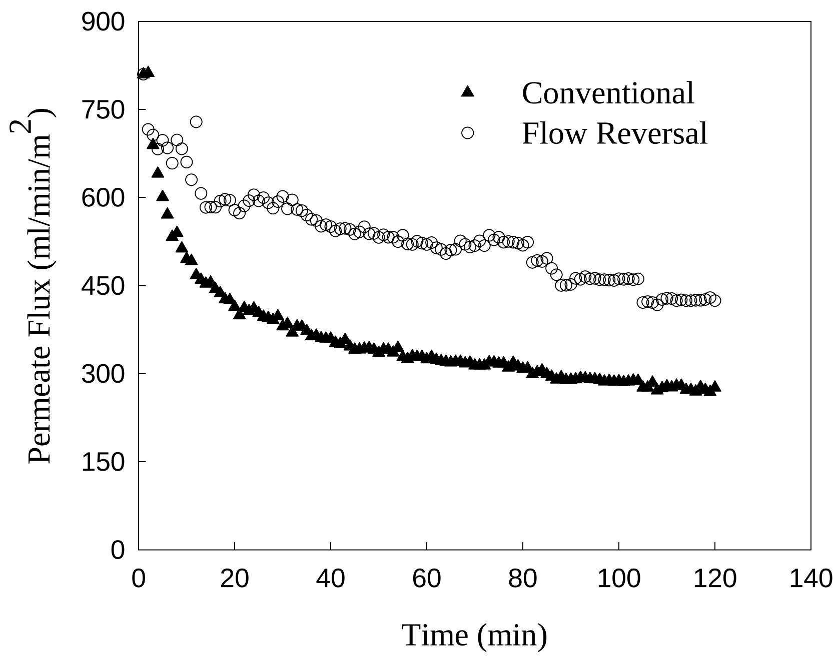


Figure 2: Comparison of permeate flux data for 0.05 wt% pectin in apple juice at a transmembrane pressure, $\Delta P_{\text{TMP}} = 30 \text{ Psia}$, flow Reynolds number, $N_{\text{Re}} = 2184,$ and flow reversal time, $\tau_{rev} = 1 \text{ min}$.

To better compare the performance of flow reversal with the conventional UF operations, permeate fluxes are plotted as flux gain against time. Flux gain is defined as the ratio of flux under Flow Reversal condition over conventional flux at a given time of operation. This is shown in Figure 3. The results clearly show that by implementing Flow Reversal technology, it is possible to achieve significant flux enhancement and the permeate flux can be maintained at a higher level for a prolonged period of time.

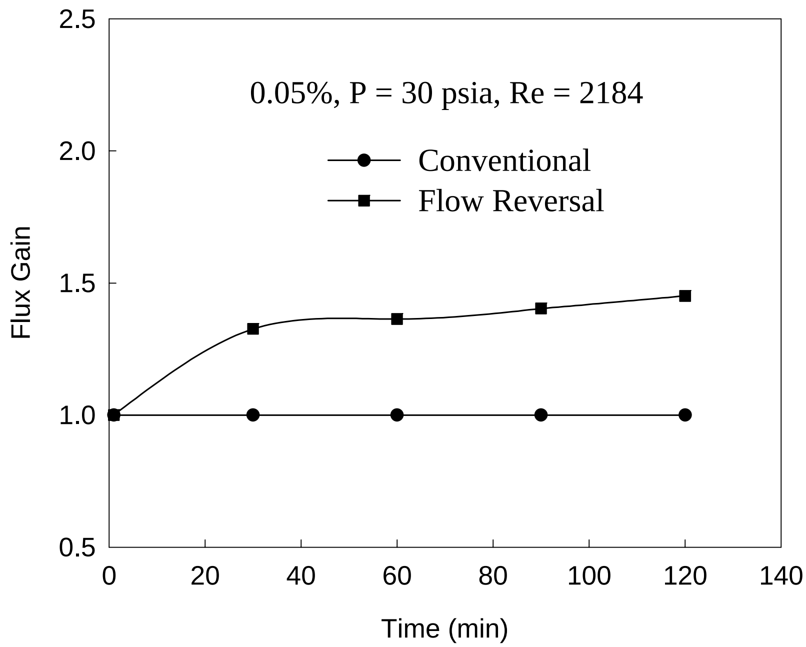


Figure 3: Flux gain curve (operating condition: 0.05 wt% pectin in apple juice at a transmembrane pressure, $\Delta P_{\mathrm{TMP}} = 30$ psia, flow Reynolds number, $N_{\mathrm{Re}} = 2184$, and flow reversal time, $\tau_{rev} = 1$ min).

With feed solution containing 0.03 and 0.01 wt% pectin in apple juice under various operating conditions we obtained similar flux enhancements with our Flow Reversal technology. Based on the experimental work to date, it appears 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 at the surface of the membrane prevents the formation of stable hydrodynamic and concentration boundary layers. 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 and results in enhanced permeate flux with the use of periodic flow reversal of the feed solution.

CONCLUSIONS

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 pectin in apple juice 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.

REFERENCES

Smolder, C.A., and van den Boomgard, Th., *J. Memb. Sci*., ***40***, 121 (1989).

Redkar, S.G., Kuberkar, V., and Davis, R.H., *J. Memb. Sci.,* ***121***, 229 (1996).

Gekas, V., and Hallstrom, B., *J. Memb. Sci*., ***30***, 153 (1987).

Cheryan, M., Ultrafiltration and Microfiltration Handbook, Technomic Publishing Company, Lancaster, Pennsylvania, 1998.

Hargrove, S., and Ilias, S., *Sep. Sci. Technol*., **34**, 1319 (1999).

Brink, L.E.S., and Romjin, J.D., *Desalination*, ***78***, 209 (1990).

Kim, K.J., Fane, A.G., and Fell, C.J.D., *Desalination*, ***70***, 229 (1988).

Ilias, S., Hargrove, S., and Talbert, M., "Flux Enhanced Crossflow Membrane Filter," U.S. Patent # 6,168,714 (2001).

Ilias, S., Hargrove, S.C., and Talbert, M., "Method for Improving the Permeate Flux of a Cross-flow Membrane Filter," U.S. Patent # 6,423,230 (2002).

Pryor, E., and Ilias, S., "Clarification of Apple Juice Utilizing a Flow Reversal Ultrafiltration Technique," 14th Annual Meeting of the North American Membrane Society (NAMS), Paper No. 401, May 17-21, 2003, Jackson Hole, Wyoming.