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

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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. 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).

Three feed solutions (Bovine serum albumin (BSA), apple juice and citrus fruit pectin) were studied in crossflow membrane filtration. These solutes are well-known in membrane filtration for their fouling and concentration polarization potentials. Laboratory-scale tests on a hollow-fiber ultrafiltration membrane module using each of the feed solutes 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. Thus, flow reversal technology seems an attractive alternative to mitigate fouling problem in crossflow membrane filtration.

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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, apple juice and citrus fruit pectin as feed in hollow fiber UF membrane module. Each of these feed solutes is well-known in membrane filtration for its fouling and concentration polarization potentials. From experimental study we observed that by flow reversal techniques, the permeate flux can be enhanced significantly by mitigating the adverse effects of concentration polarization and fouling in crossflow membrane filtration. Thus, the concept of periodic flow reversal of feed flow in crossflow UF operation provides an option for enhanced productivity by overcoming the limitations of concentration polarization and 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 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 permeate 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.

The major emphasis in the design and operation of crossflow 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 [Bruin, et al., 1980; Ilias and Govind, 1990; Belfort, et al., 1994]. Some of the popular schemes that have been practiced or are being considered for flux enhancement in cross flow filtration are shown in Figure 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 Figure 1(a). With a rapid drop in pressure, transmembrane flux also drops rapidly with time. The problem is complicated by the fact that if one uses high inlet pressure, 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 under utilized as shown schematically by the performance curve in Figure 1(a). To overcome these limitations, periodic reversal 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 Figure 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 [Jonsson, 1993].

An improved version of lumen flush is the periodic backwash (PBW), which is conducted by pumping the permeate at higher pressure across the membrane to the feed side. This result in lifting or dislodging deposited materials from the membrane surface. As shown in Figure 1(c), PBW can provide higher flux but its effectiveness may decrease with time especially if pore fouling is the main cause [Rodgers and Sparks, 1993]. 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 co-current permeate flow (CPF). As shown in Figure 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 Figure 1(d). The

UTP/CPF has been credited for enhanced flux in crossflow UF and MF operations [Gesan et al., 1995].

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 [Ilias et al., 2001; Hargrove, 1998; Ilias et al., 2002]. 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. From limited UF experiments with BSA, we observed that flow reversal significantly improves the permeate flux and merits further research.

In this project, 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, apple juice and citrus fruit pectin as feed solutes with and without periodic flow reversal option and compare performances.

EXPERIMENTAL: MATERIALS & METHODS

Cross flow membrane filtration experiments were conducted in tubular UF membrane modules using bovine serum albumin (BSA), apple juice and citrus fruit pectin as feed solutions. Each of these feed solutes is well-studied in membrane filtration, and is known for its potent fouling and concentration polarization capabilities.

BSA: 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.

Apple Juice: Apple pectin from the Sigma Aldrich Company was used as the major foulant in the feed solution. Product specifications: Poly-D-galacturonic acid methyl ester, product specification number P-8471, lot number 030K0883, degree of esterification 60 – 75 %. In all experiments store bought apple juice was used to simulate a pretreated clear juice. Apple pectin solutions were prepared in apple juice at various concentrations of pectin (0.01-0.05 wt%) before each experiment was conducted.

Citrus Fruit Pectin: Citrus Fruits Pectin (CFP) powder was purchased from Sigma Aldrich Company, the product specification number is P-9135 and the Lot number is 032K1258. The product contain 93.5% (as is) Galacturonic, Methoxy content 9.4% (as is), and loss in drying about 7.3%. The deionized water was prepared using a corning MEGA-PURE deionization filtration system. According to the manufactures specifications, this system uses Ultra-High Purity disposable deionizer cartridges and delivers reagent-grade water that suitable to use in ultrafiltration membrane separations.

Membrane Modules:

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 2. The forward feed flow and the reverse feed flow schemes are shown here. The forward feed flow scheme (Figure 2(a)) is the one that is commonly used in cross flow membrane filtration operation. Reverse flow mode is shown in Figure 2(b). 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.

In order to facilitate reversal of direction of flow of the feed stream within the membrane module a Fisher Brand TM lab controller and a custom-made actuated elliptic valve system were used. The actuated valve was specially ordered from Cole-Parmer Instrument Company and is stacked 2 high. The valve operates on a 90° angle valve actuation whose cycle time is approximately 2.5 seconds.

The feed solution is recirculated using a Progressive cavity pump, fitted with a DC speed control and a programmable tachometer. The permeate flow rate is sensed by the digital flow sensors acquired from CTE (ChemTec company). The analog voltage outputs from the sensors are connected in parallel to the digital panel meters (Cole Parmer Instrument Company) and the data acquisition card (KPCI-3101) from Keithley instruments. The LabVIEW program developed for the data acquisition enables real time visual permeate flow rate characteristics under the operating conditions. Other supporting equipments essential for the experiment are pressure gauges, and back pressure valve.

Pure water flux data was collected prior to using a new membrane. Membranes were cleaned following each experiment, and pure water flux data was taken again after cleaning and the water flux was normalized for both the system (flow reversal and unidirectional). The flux regeneration procedure for the membrane is as follows:

- o Cleaning with water at 50 °C for about 4 hrs.
- o Cleaning with 0.5 N Sodium hydroxide solutions at 50 °C for about 2 hrs.
- o Cleaning again with water for about 5-6 hours.
- o Cleaning with water at 50 °C till the flux is regenerated to about the original value.

A photograph of the laboratory-scale flow reversal membrane filtration unit in operation is shown in Figure 3. The PC is used to run LabVIEW for data acquisition and controlling the UF operations.

LabVIEW Data Acquisition System

It comprises of the following:

- o Flow sensors with analog output of 0-5 VDC.
- Digital panel meters (Cole Parmer Instruments Company): Accepts input from 0-5 VDC.
- ended or pseudo-differential analog input channels, or 8 differential analog input channels on board. The KPCI-3101 series board can acquire data from a single analog input channel or from group of analog input channels. Onboard channels are numbered (0-15) for single-ended and differential input channels or (0-7) for differential inputs.
- LabVIEW 6i National Instruments: LabVIEW is a fully featured graphical programming language used to create virtual instrumentation. It consists of an interactive user interface, complete with knobs, slide switches, graphs, strip charts, and other instrument panel controls. The function blocks, which are selected from palette menus, range from arithmetic functions to advanced acquisition, control, and analysis routines. Also included are debugging tools, help windows, execution highlighting, single stepping, probes and breakpoints to trace and monitor the data flow execution. A Keithley VI palette provides standard virtual instruments (VIs) for LABVIEW that interface with KPCI-3101 board through Driver LINX.
- Driver LINX software provides interface to configure analog and digital I/O modes without Register level programming.

The schematic of the Data Acquisition set up is shown in Figure 4.

- o Install the LabVIEW software.
- o Installing the KPCI-3101 board.
- o Turn of the computer and all its peripherals.
- o Select the 32-bit or 64-bit PCI expansion slot.

- After physically installing the board, turn on and reboot the computer. Run the Driver LINX wizard.
- Before making any connections to the board, check whether Driver LINX and Board are installed correctly and working together properly.
- o Click on the Driver LINX Analog I/O panel.
- o The analog I/O panel is useful for testing the KPCI-3101-4 series board Driver LINX installation and configuration, verifying the signal inputs to the PCI board, sending test signals top external devices, controlling the DC output voltages of two output channels, setting and reading all digital input and output bits on the board.
- o Attach the STP-68 screw terminal panel.
- Attach the 68-pin connector to the KPCI-3101 board using the CAB-305 cable. There
 is one to one correspondence between pins and terminals.

The LabVIEW program developed for the data acquisition is shown in Figure 5. The LabVIEW program comprises of three modules: the do-while loop for the real time plot, a for-loop for totalizer or flow accumulation and a for-loop for saving the data automatically to an excel-spread sheet. In all these loops, the program acquires the real time computer clock and uses it for the data calculation and calibration. In addition the program also controls the rate of data acquisition.

RESULTS AND DISCUSSIONS

Cross-flow membrane filtration experiments were performed in polysulfone UF tubular membrane modules with BSA, apple juice and citrus fruit pectin as feed solutions. The performance of the UF fluxes for the three feeds are presented here.

BSA Solution:

The BSA feed concentration ranged from 0.01 wt% to 5 wt% and the operating transmembrane pressure ranged from 20 Psia to 30 Psia. Trans-membrane permeate flux data was collected for both the unidirectional and flow reversal conditions. For comparison purpose, 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 Figures 6 and 7, respectively. The data shows that there is noticeable gain in permeate flux with flow reversal. A comparison of the flux data in Figures 6 and 7 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 two minutes, i.e., in every two minutes the direction of the feed and permeate flows were reversed using the computer controlled valve manifolds. The flow switching time of two minutes was chosen because the flux decline in crossflow 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 short interval of time. This helps in minimizing 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 crossflow filtration.

Figures 8 and 9 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) crossflow filtration at higher transmembrane pressure. This is supported by the experimental flux data in Figures 8 and 9 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/m² in about one hour of UF operation (Figure 8). With flow reversal, the permeate flux can be maintained at about 200 ml/min/m² for a prolonged period of time.

Based on the experimental results presented above, 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 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.

Apple Juice:

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. The pectin content in the apple juice ranged from 0.01 wt% to 0.05 wt% and the operating transmembrane pressure $\Delta P_{\rm TMP}$ ranged from 20 Psia to 30 Psia. Trans-membrane permeate flux data was collected for both the unidirectional and flow reversal conditions. Effect of feed flow rates and flow reversal times on permeate flux were investigated.

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.

In Figures 10 through 12, the permeate flux and flux gain data presented three feed concentrations. Flux gain is defined as the ratio of flux under Flow Reversal condition over conventional flux at a given time of operation. The operating transmembrane pressure difference was 30 Psia and feed flow Reynolds number was, $N_{\rm Re}=2184$. With conventional UF operation, permeate flux drops rapidly with time. For the same operating conditions using our flow reversal technology, with flow reversal time, $\tau_{\rm rev}=1\,{\rm min}$, we find a marked improvement in transmembrane flux

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.

Citrus Fruit Pectin:

Cross-flow ultrafiltration experiments were performed with CFP as a feed solution over a concentration range of 0.03 wt% to 0.1 wt% and at feed velocities of 1.0 m/s, 1.2 m/s and 1.41 m/s. Data was collected for both conventional and reversal conditions, with conventional flow as the base case. Each experiment was conducted for at least 90 min with flow reversal and/or data collection each minute. The main objective was to understand the permeate flux behavior of CFP under various operating parameters, such as feed concentration, flow reversal time, feed velocity, and feed transmembrane pressure.

The effect of feed concentration on flow reversal was studied at CFP feed concentrations of 0.03 wt%, 0.05 wt5, 0.07 wt5, and 0.1 wt% CFP at a feed velocity of 1.2 m/s and TMP of 40 psi. Figures 13 to 16 show the variation of permeate flux with time, with and without flow reversal technique. The data show that flow reversal provides a higher permeate flux when compared to the conventional without flow reversal case. As mentioned earlier, the membranes used in this study are polysulfone UF membranes. Polysulfone membranes are hydrophilic. It is common to see a characteristic permeate flux rise when hydrophilic membranes are used in the separation of aqueous protein solutions. Since hydrophilic membranes show high protein adsorption, a distinct layer of absorbed protein develops as the membrane is wetted, thus changing the membrane's properties. The additional protein layer causes the membrane to behave more like a hydrophilic membrane and the flux performance then follows the trend normally observed with hydrophilic membranes [Baker, 2000]. Therefore, it is expected that a more sizeable flux will occur for more dilute solutions. The increase is more pronounced when flow reversal is used because the increased hydrodynamic instability slows protein adsorption on the membrane's surface.

Figure 17 shows that flow reversal technique has a positive impact on permeate flux even at a higher concentration which implies that flow reversal effectiveness would vary at a different concentration. Flow reversal is also more effective at a lower concentration of the feed solution as the solute deposition on the membrane surface decreases on reducing the concentration. Examination of the results from Figure 17 shows clearly that with the increasing CFP concentration at a constant feed velocity and transmembrane pressure, the permeate flux in crossflow UF diminishes. The average increases in permeate flux when using flow reversal at 0.03 wt%, 0.05 wt%, 0.07 wt% and 0.1 wt% are 11.03%, 13.9%, 15.03%, and 16.04% respectively.

The overall average increase in permeate flux at a different concentration is 14.2% which is higher compared to 9.45% that was reported by Hargrove (1998) using a different concentration of BSA solution. The difference in permeate average between the two studies is due to the different concentrations used in both experiments. Despite the decrease in permeate flux, a definite trend of greater flux with flow reversal than without flow reversal emerges.

Flow reversal time is a very important factor when investigating the behavior of permeate flux under flow reversal condition. It is critical to disturb the steady development of the resistance layer. Figure 18 is a comparison of permeate flux data. The figure shows the effects of flow reversal time ($\tau_{rev} = 1 \text{ min}, 2 \text{ min}, \text{ and } 3 \text{ min}$) on permeate flux under flow reversal technique for 0.05 wt% CFP solution at transmembrane pressure of 40 psia, and at a feed velocity of 1.2 m/s. The average increase in permeate flux were found to 13.03%, 7.42% and 3.35% for flow reversal times of 1 min, 2 min and 3 min, respectively. The figure clearly shows that the flow reversal time of 1 min is a very effective flow reversal process and it seems to enhance the permeate flux significantly for this particular feed.

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 BSA, pectin in apple juice and citrus fruit pectin as feed solutions. 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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- 3. Parthasarathy, H., "Flux Enhancement in Crossflow Membrane Filtration: Fouling and Its Minimization by Flow Reversal," M.S. Thesis, North Carolina A&T state University, 2005.

REFERENCES

Baker, R., Membrane technology and applications. McGraw-Hill, New York, 2000.

Belfort, G., Davis, R.H., and Zydney, A.L., J. Memb. Sci., 96, 1 (1994).

Bruin, S., Kikkert, A., Weldring, J.A.G., and Hiddink, J., Desalination, 35, 223 (1980).

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

Cheryan, M., <u>Ultrafiltration and Microfiltration Handbook</u>, Technomic Publishing Company, Lancaster, Pennsylvania, 1998.

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

Gesan, G., Daufin, G., and Merin, U., J. Memb. Sci., 104, 271 (1995).

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

Hargrove, S.C., M.Sc. ChE Thesis, North Carolina A&T State University, Greensboro, NC, 1998.

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).

Ilias, S., and Govind, R., Sep. Sci. Technol., 25, 1307 (1990).

Jonsson, A.S., J. Memb. Sci., 97, 37 (1993).

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

Rodgers, V.G.J., and Sparks, H.E., J. Memb. Sci., 78, 163 (1993).

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).

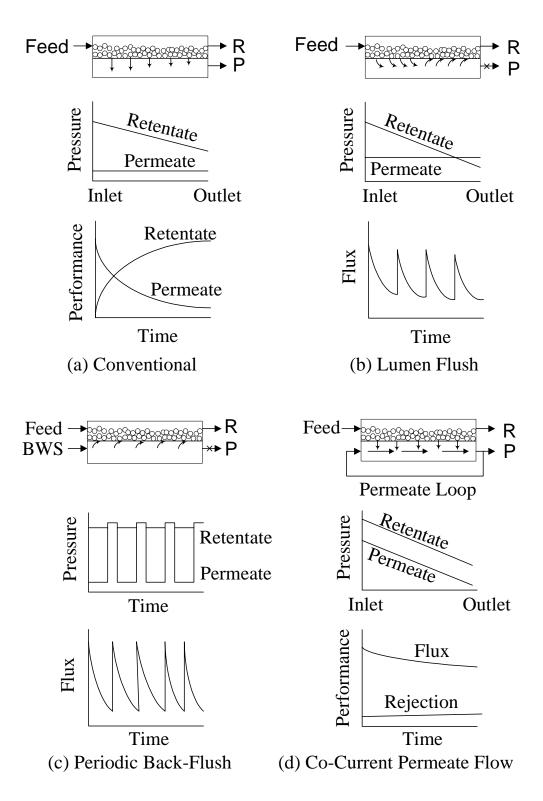
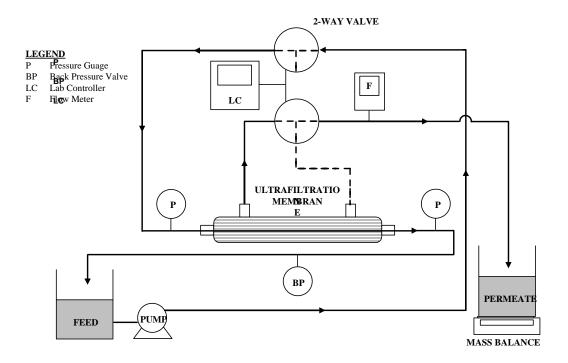


Figure 1: (a) Conventional, (b) Lumen Flush, (c) Periodic Back-wash, and (d) Co-current Permeate Flow schemes in cross flow membrane filtration, showing expected behavior of pressure profiles and time-dependent flux (P = Permeate, R = Retentate, and BWS = Back-Wash Solvent).



(a) Forward Feed Flow

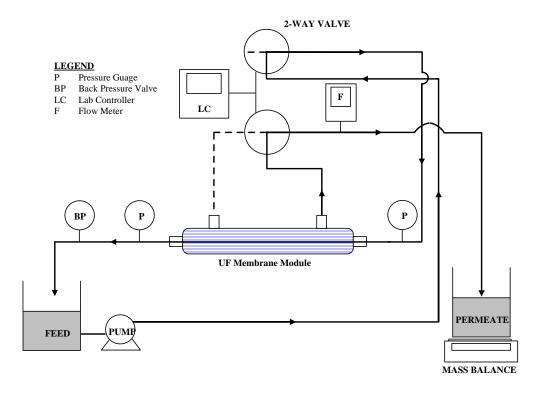


Figure 2: Schematic of the experimental set up. (a) Forward Feed Flow, and (b) Reverse Feed Flow mode of operations.



Figure 3: Photograph of prototype flow reversal cross flow membrane filtration unit.

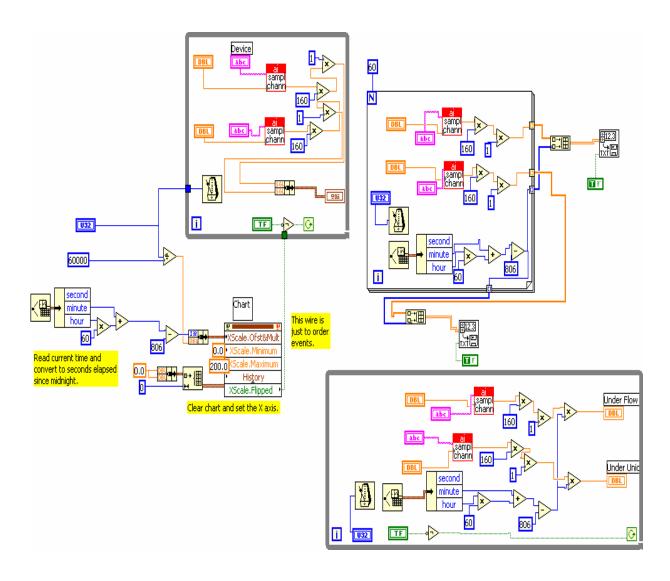


Figure 4: Data Acquisition program in LabVIEW 6i.

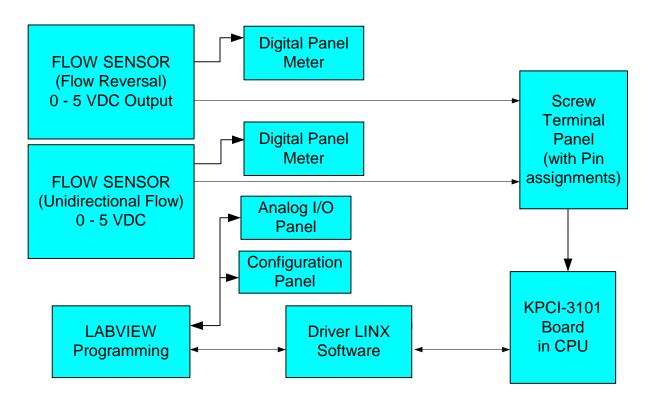


Figure 5: Data acquisition scheme for the membrane filtration process.

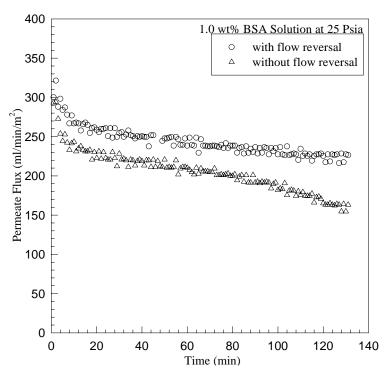


Figure 6: Comparison of permeate flux data for 1.0 wt% BSA solution at a transmembrane pressure of 25 Psia.

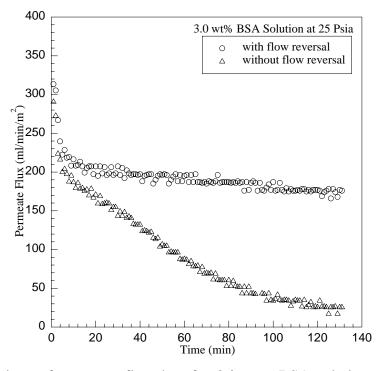


Figure 7: Comparison of permeate flux data for 3.0 wt% BSA solution at a transmembrane pressure of 25 Psia.

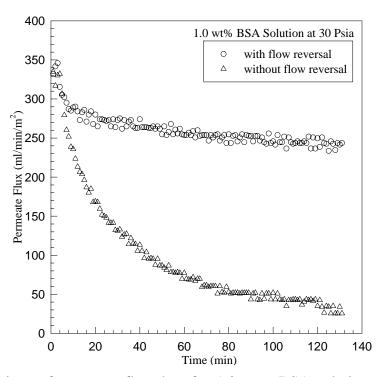


Figure 8: Comparison of permeate flux data for 1.0 wt% BSA solution at a transmembrane pressure of 30 Psia.

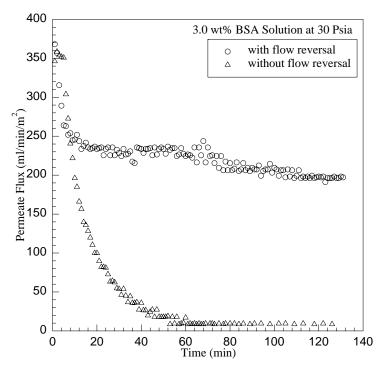


Figure 9: Comparison of permeate flux data for 3.0 wt% BSA solution at a transmembrane pressure of 30 Psia.

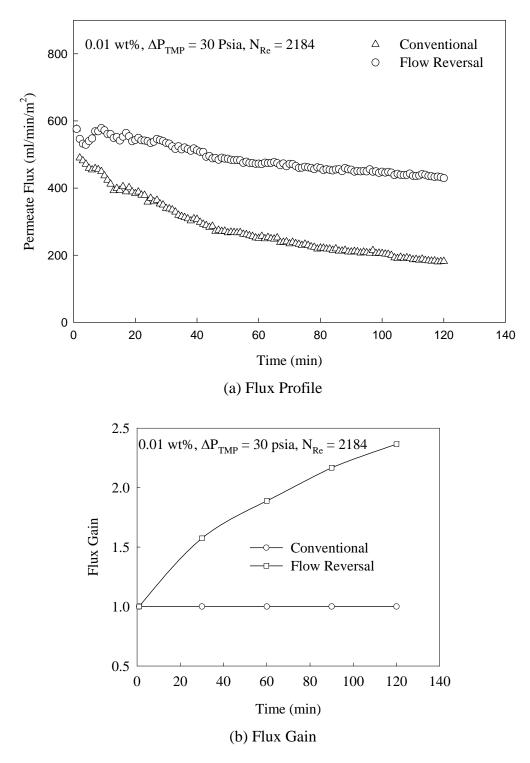


Figure 10: Comparison of (a) permeate flux data and (b) flux gain for 0.01 wt% pectin in apple juice at a transmembrane pressure, $\Delta P_{\rm TMP}=30$ Psia, flow Reynolds number, $N_{\rm Re}=2184$, and flow reversal time, $\tau_{\rm rev}=1$ min.

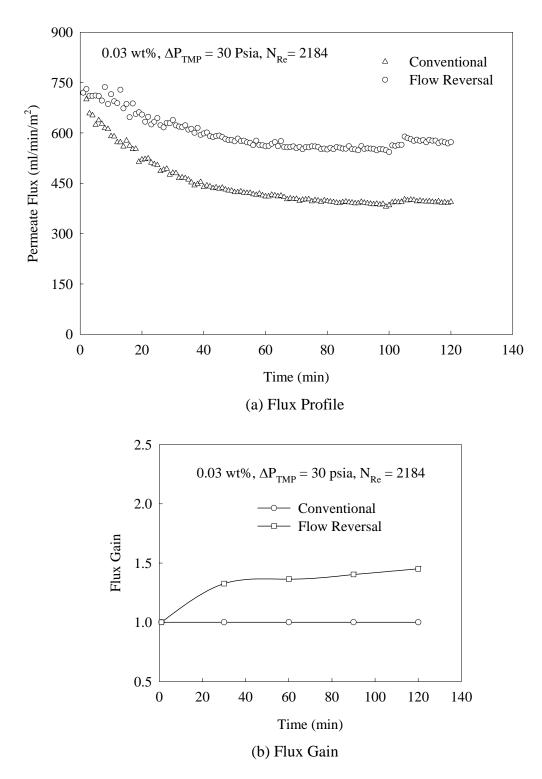


Figure 11: Comparison of (a) permeate flux data and (b) flux gain for 0.03 wt% pectin in apple juice at a transmembrane pressure, $\Delta P_{\rm TMP} = 30$ Psia, flow Reynolds number, $N_{\rm Re} = 2184$, and flow reversal time, $\tau_{\rm rev} = 1$ min.

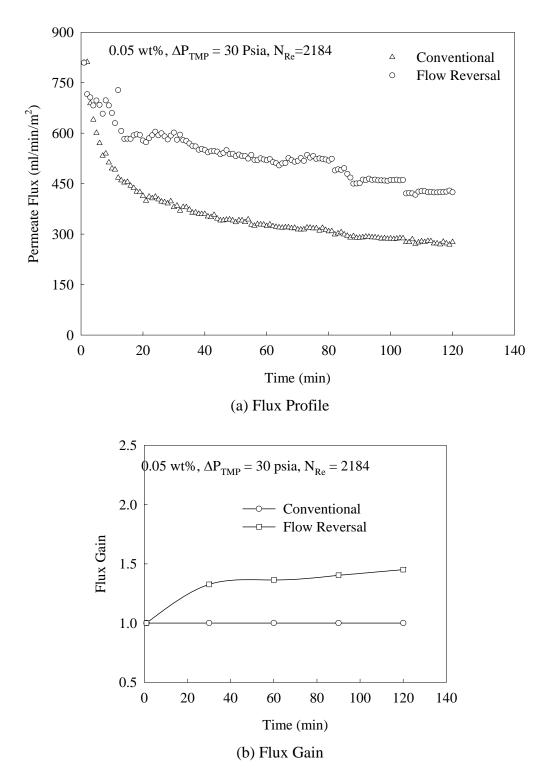


Figure 12: Comparison of (a) permeate flux data and (b) flux gain for 0.05 wt% pectin in apple juice at a transmembrane pressure, $\Delta P_{\rm TMP} = 30$ Psia, flow Reynolds number, $N_{\rm Re} = 2184$, and flow reversal time, $\tau_{\rm rev} = 1$ min.

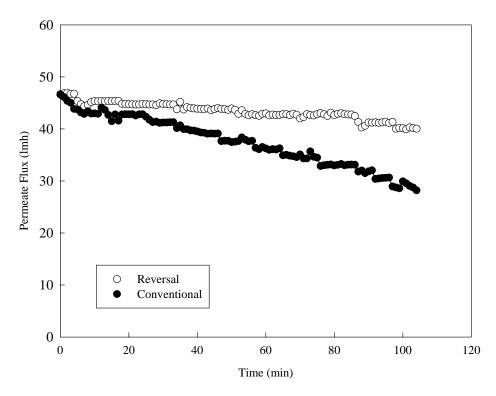


Figure 13: Comparison of permeate flux data for 0.03 wt% CFP solution at transmembrane pressure of 40 Psia, and feed velocity of 1.2 m/s.

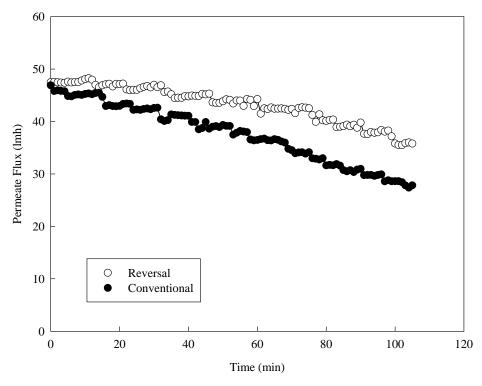


Figure 14: Comparison of permeate flux data for 0.05 wt% CFP solution at transmembrane pressure of 40 Psia, and feed velocity of 1.2 m/s.

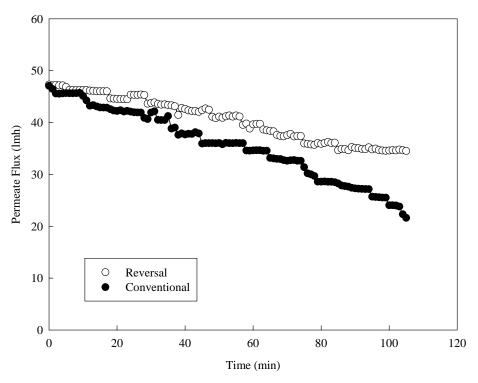


Figure 15: Comparison of permeate flux data for 0.07 wt% CFP solution at transmembrane pressure of 40 Psia, and feed velocity of 1.2 m/s.

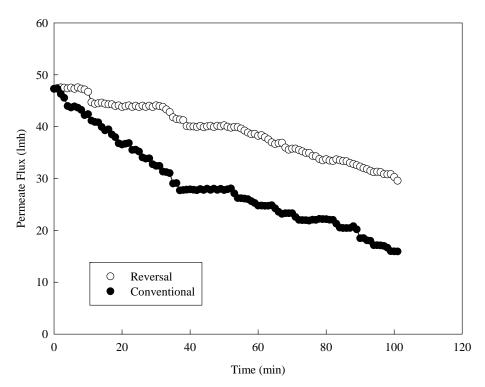


Figure 16: Comparison of permeate flux data for 0.1 wt% CFP solution at transmembrane pressure of 40 Psia, and feed velocity of 1.2 m/s.

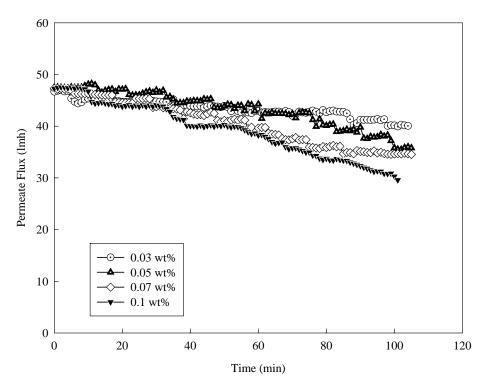


Figure 17: Comparison of permeate flux: Effect of concentration on permeate flux under flow reversal for CFP solution at transmembrane pressure of 40 Psia, and feed velocity of 1.2 m/s.

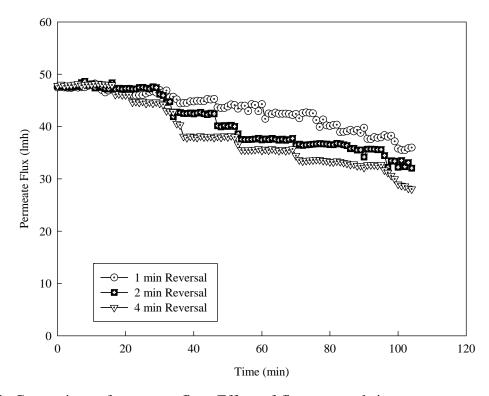


Figure 18: Comparison of permeate flux: Effect of flow reversal time on permeate flux under flow reversal for 0.05 wt% CFP solution at TMP of 40 psi, and feed velocity of 1.2 m/s.