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FLUX ENHANCEMENT USING FLOW REVERSAL IN ULTRAFILTRATION

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ABSTRACT

The effect of flow reversal on permeate flux in cross-flow ultrafiltration of bovine serum albumin (BSA) has been investigated experimentally. BSA is a well-studied model solute in membrane filtration known for its fouling and concentration polarization capabilities. Ultrafiltration experiments were performed with BSA feed solutions in a hollow-fiber membrane module. The BSA feed concentrations ranged from 0.01 to 5 wt% and were ultrafiltered at a transmembrane pressure of 20 psia. Permeate flux was determined both with and without the use of flow reversal for each concentration. The experimental results indicate that under flow reversal conditions, the permeate flux is enhanced significantly when compared with runs without flow reversal. The effect of flow reversal on flux enhancement is very pronounced for dilute BSA solutions.

INTRODUCTION

The existence of most substances as solutions or mixtures created the need for processes to be developed to separate these solutions or mixtures. In this context, the need to purify, recover, isolate, and remove substances in process streams in chemical, pharmaceutical, food, petroleum, and wastewater applications has driven investigations in separation technology. In recent years, membrane-based cross-flow filtration has gained importance in many separation applications and, in some situations, competes with traditional separation technologies, such as distillation, absorption, and extraction (1).

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The most common cross-flow membrane processes are microfiltration (MF), ultrafiltration (UF), and reverse osmosis (RO). Although varying in transmembrane pressure difference driving force and average pore diameter, each membrane serves as a selective barrier by permitting certain components of a mixture to pass through while retaining others. This results in two phases, permeate and retained phases, each of which is enriched in one or more of the components of the mixture.

The major resistances to the successful use of membrane separation processes are phenomena known as *concentration polarization* and *fouling*. Concentration polarization occurs when a concentration gradient of the retained components is formed on or near the membrane surface (2). Fouling is the deposition of material on the membrane surface or in its pores, leading to a change in membrane behavior or even pluggage (2). These phenomena manifest themselves in that with time and increased operating pressure, the permeate flux reaches an asymptotic value beyond which further increases in operating time and pressure do not result in increased flux. The severity of the effects of these phenomena varies with the membrane type and the composition of the process stream.

Concentration polarization is a function of the hydrodynamic conditions in the membrane system (3). Membrane fouling is usually characterized as irreversible; however, when cross-flow membranes are used, the imposed stress of the cross-flow tends to shear the foulant layer. Hence, varying the fluid mechanics of a system is very important in maximizing the total capacity of a membrane module (4). In the past, a number of investigators attempted manipulation of fluid hydrodynamics or membrane surface morphology to enhance transmembrane flux, with limited success (4,5).

The effect of membrane surface modification by chemical and physical means has been investigated. The principle behind the idea of chemical modification is that it might reduce attractive forces or increase repulsive forces between the solute and membrane. However, this method has been found to have little effect on the behavior of suspended particles once a secondary cake has been established (4). Physical modification is achieved by using protuberances designed to induce instabilities in the bulk flow. Protrusions are actually placed on the membrane surface at defined intervals in such applications; therefore, useful surface area is diminished. Additionally, this technique causes high axial pressure drops and is difficult to scale up (4).

Techniques to modify fluid hydrodynamics of the bulk stream have also been investigated. A study of flow pulsation by periodic induction of a pressure gradient on the feed stream focused on flux enhancement and power consumption (5). It has been found

that the advantage of using pulsation supersedes the disadvantage of increased power consumption. However, the problems of energy dissipation and reduced cross-flow, which results in a lower net filtering capacity, remain (4). Fluid instabilities due to flow in curved ducts, known as Taylor and Dean flows, have been used to disturb the flux-limiting effects of concentration and fouling (6). The problems associated with this and other external devices similar to it are the high energy required to operate the devices, the difficulty to repair them, and the difficulty to scale them up (4).

We are currently investigating 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. We have chosen to investigate the UF of bovine serum albumin (BSA) in aqueous solution as a model feed. Interpretive studies have shown that in the UF of BSA both concentration polarization and deposition on the front face of the membrane as foulant are important (3). We are proposing to change the hydrodynamics of typical membrane systems by periodically reversing the direction of flow of the feed stream in the membrane. Periodic reversal of the direction of flow of the feed stream in the membrane module, while maintaining the cross-flow, keeps the system in a hydrodynamically transient state and prevents the formation of a stable boundary layer at the membrane surface. Therefore, the collection of particles in a gradient near the membrane surface and particle deposition on the membrane surface are slowed. Our flow-reversal technique should not be confused with backpulsing, in which the permeate stream is periodically forced back through the membrane under the impetus of an induced pressure gradient. The objective of this paper is to report some preliminary results to show the potential application of flow reversal as a flow manipulation technique to enhance membrane fluxes.

MATERIALS AND EXPERIMENTAL METHOD

The BSA solutions were prepared by dissolving appropriate amounts of Bovine Albumin Fraction V Powder in distilled water. Complete solution homogeneity was facilitated by stirring each sample on a stir plate until no solids were present in the liquid. Each solution was freshly prepared immediately before each experimental run. For these preliminary experimental runs, the pH of the feed solution was not controlled by adding

any buffers. To cover a wide range of feed concentrations, 0.01, 0.1, 0.5, 1, and 5 wt% BSA solutions were used.

Experiments were conducted using a polysulfone UF membrane with a nominal molecular weight cut-off of 3000, manufactured by A/G Technology. The membrane module has an effective length of 12.4 in, and contains 13 fibers, each with an internal diameter of 1 mm. The performance of the membrane was maintained by following the manufacturer's cleaning procedure. The cleaning procedure includes flushing with clean water, buffer, or saline solution at 50°*C*; circulation of 0.5 *N* NaOH solution at 50°*C* for 1 h, and a final flushing with clean, warm water.

The effectiveness of this procedure was evaluated by determining the pure water flux over a 10-min period both prior to and after each experimental run. For each comparison, the pure water flux data were comparable within 10% with occasional improvements in the membrane performance. These fluctuations in the membrane performance can be attributed to slight temperature changes in the membrane water bath and some membrane compaction over time. By the time the final pure-water flux data were taken, the membrane had deteriorated slightly less than 12.5%.

A schematic drawing of the experimental setup is shown in Figure 1. The BSA solutions are circulated through the experimental apparatus using a piston, positive-displacement pump manufactured by Fluid Metering, Inc. This pump is equipped with a variable-speed pump head and controller to vary the flow rates. The feed stream then flows into the valve. A controller regulates the valve position. For these experiments, the controller was set to change the valve position every minute. The valve directs the flow of the membrane feed and permeate streams. From the valve, the stream enters the membrane on the axial ports. The permeate stream exits the membrane from the radial ports. The driving force of this separation is the transmembrane pressure. Here, a pressure relief valve is used to regulate the pressure and the corresponding pressure is measured by pressure gauges at each axial port. The retained phase then recycles back to the feed reservoir. The permeate stream is collected and its mass recorded each minute.

Following each experimental run, a sample was taken from the permeate reservoir for analysis to determine the remaining BSA concentration following UF. Sigma Diagnostics Procedure No. 631 was used. In short, each sample was mixed with Sigma Albumin Reagent (BCG) to facilitate a color change, and a spectrophotometer was then

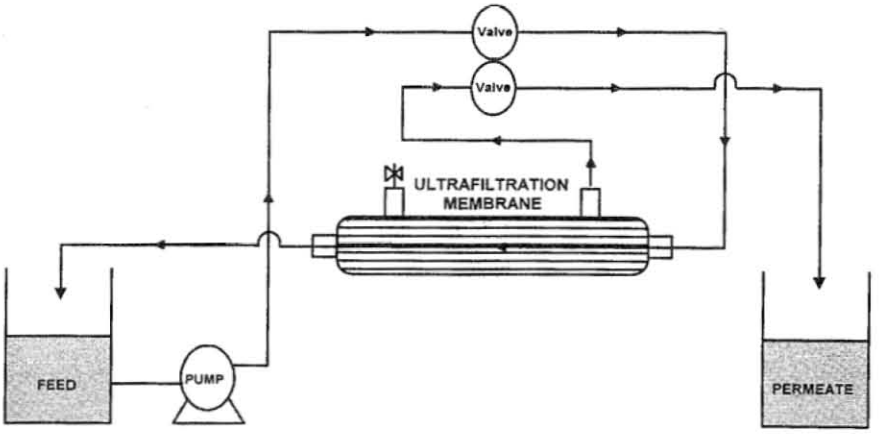

FIGURE 1. Simple schematic of the experimental setup.

used to determine the absorbance of the sample at a wavelength of 628 nm. Absorbances were also determined for a "blank" using distilled water and a standard sample of a BSA solution of known concentration. The albumin concentration of the sample was then determined as

$$C_s = \frac{A_s - A_b}{A_{ref} - A_b} \times C_{ref} \tag{1}$$

where C_i is the concentration, in wt %, with subscripts *s* for sample and *ref* for standard solution; and A_i is the absorbance, with subscripts *s* for sample, *b* for blank, and *ref* for standard reference solution, respectively.

The rejectivity (%) of BSA by the UF membrane was calculated using the following equation:

$$\beta = \left(1 - \frac{C_p}{C_0}\right) \times 100 \tag{2}$$

where β is the solute rejectivity of the membrane, and C_o and C_p are the concentrations of the feed and permeate (wt %) solutions, respectively.

RESULTS AND DISCUSSION

Cross-flow ultrafiltration experiments were performed with BSA as feed solution over a concentration range of 0.01 wt % to 5 wt % under flow reversal and unidirectional flow conditions. A single membrane module was used for the experiments reported here. Before each experimental run, the membrane module was thoroughly cleaned to restore the membrane. Flux data for pure water were then collected and compared with the initial data. We found that there was a gradual decline in the flux for pure water over a long period of operation. Each of the ultrafiltration experimental runs with and without flow reversal were conducted for slightly more than 2h to study the transmembrane flux behavior.

Figure 2 shows the variation of permeate flux with time, with and without flow reversal, for 0.01 wt % BSA feed solution. This is the lowest feed concentration used in our study. The data show that flow reversal provides a higher permeate flux when compared with the case without flow reversal. One interesting behavior worth noting in both cases is the surge in permeate flux during the first 20 to 30 min. This is in agreement with the findings reported by other workers (7) and can be explained in terms of membrane properties. As mentioned earlier, we used a polysulfone UF membrane, which is known to be hydrophobic. When using hydrophobic membranes to separate aqueous solutions, there is a characteristic permeate flux rise that is attributed to membrane properties. Since hydrophobic membranes show high protein adsorption, a distinct layer of absorbed protein develops as the membrane is wetted, thus changing the membrane properties. The protein layer causes the membrane to behave more like a hydrophilic membrane, and flux performance follows the trends normally observed with such membranes (7). Therefore, it is expected that a more sizable flux increase 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 surface. Nonetheless, there is an average increase of 4.6% in permeate flux when flow reversal is used.

In Figure 3, the variation of permeate flux with time for a 0.1 wt % BSA feed solution is shown for the cases of UF with and without flow reversal. The data show that there is a noticeable gain in permeate flux with flow reversal, 13.3% on average. In both

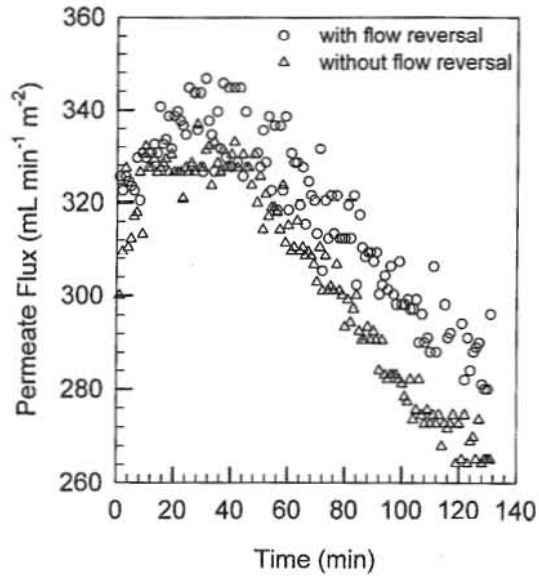

FIGURE 2. Comparison of permeate flux data for 0.01 wt % BSA.

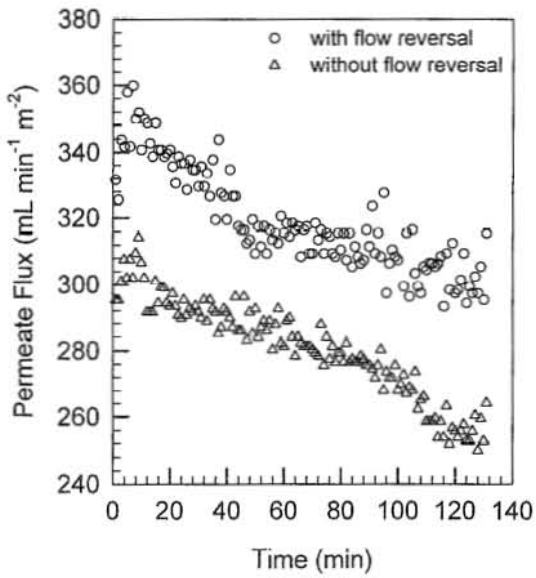

FIGURE 3. Comparison of permeate flux data for 0.1 wt % BSA.

cases, the flux declines with time. However, in the case of flow reversal, the rate of decline is much slower than in the case without flow reversal. It is also to be noted that there is a similar surge in permeate flux during the first 10 to 15 min in both cases. As stated above, this behavior is in agreement with results reported by others (7).

Figures 4–6 show the permeate flux data with time for higher BSA feed concentrations. As shown in Figure 4, for a BSA concentration of 0.5 wt % we observed an initial increase in permeate flux for about 5 min in the case of UF with flow reversal. This behavior was not observed in the case of UF without flow reversal. In both cases, flux declined with time but there was a rapid decline in flux when flow reversal was not used. On average, there was a 17.4% increase in permeate flux when flow reversal was used. With increasing BSA concentration in the feed, the effect of flow reversal on permeate flux in UF diminished. This is shown in Figures 5 and 6 for 1 and 5 wt % of BSA in the feed, where average flux increases were 7.2 and 11.7%, respectively. However, a definite trend emerged from the results, the permeate flux was greater when flow reversal was used. The UF results for 1 wt % BSA solution are shown in Figure 5. The data for the case of flow reversal exhibit unique behavior. The permeate flux for the two cases is essentially equal during the first 55 min. After about 1h the flux with flow reversal dramatically surges upward to give an increased flux characteristic of our other data. The closeness of the initial data points lowered the average percentage difference, which we expected to be nearer 13%. We are currently unable to explain this behavior.

The percentage gain in permeate flux with the use of flow reversal for each BSA concentration follows a definite trend, as seen in Figure 7. It appears that flow reversal has a window of greatest effectiveness from approximately 0.1 to 1 wt %. Ultrafiltration of solutions with compositions outside this range can still benefit from flow reversal, although not as significantly.

Each of the graphs in Figures 2–6 is characterized by a gradual systematic decline in permeate flux. This behavior has been observed and confirmed by several researchers (8,9). It is believed that a number of phenomena acting simultaneously reduce the permeate flux. Accordingly, in the first minute, the initial rapid drop in flux is due primarily to concentration polarization. The flux then continues to decline, initially rapidly, for up to 1 h due to protein deposition (8). Then, a quasi-steady-state period is reached where the flux declines slowly, possibly due to further deposition of particles or to consolidation of the fouling layer (9). This is shown schematically in Figure 8.

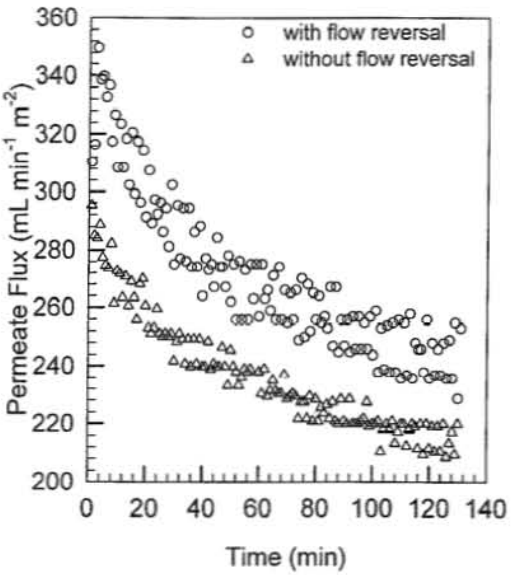

FIGURE 4. Comparison of permeate flux data for 0.5 wt % BSA solution.

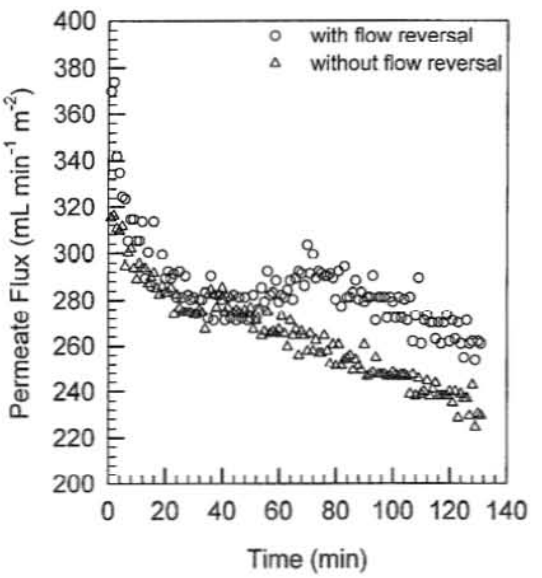

FIGURE 5. Comparison of permeate flux data for 0.5 wt % BSA solution.

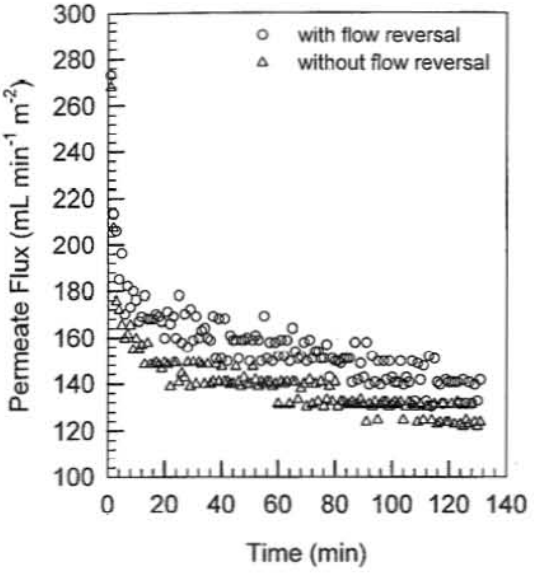

FIGURE 6. Comparison of permeate flux data for 5 wt % BSA solution.

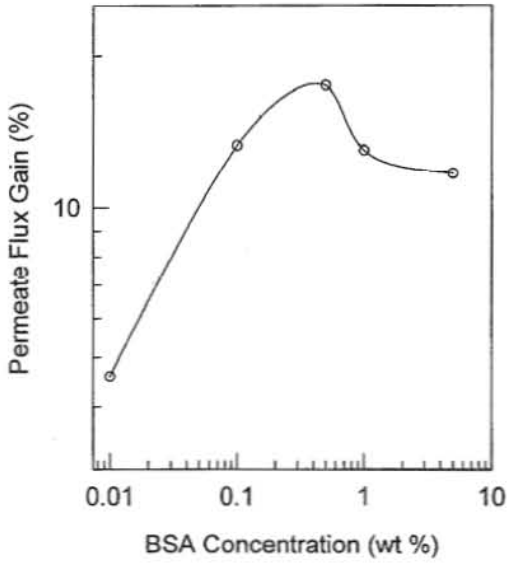

FIGURE 7. Average percent gain in permeate flux when flow reversal is used.

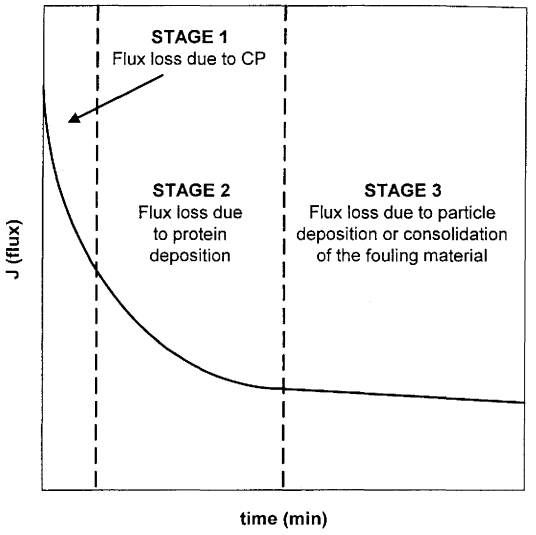

FIGURE 8. Stages of flux decline.

Based on this information and the experimental results presented in this work, it appears that flow reversal mitigates the effects of the concentration polarization that cause the initial rapid decline in permeate flux. This can be further explained using the concentration- and gel-polarization concepts (1). The gel-polarized layer shown in Figure 9 is a result of protein build up in the hydrodynamic boundary layer at the surface of the membrane. Periodic reversal of the direction of the feed stream at the surface of the membrane prevents the formation of a stable hydrodynamic boundary layer. However, as the UF progresses over time and protein macromolecules are retained by the membrane, some protein adsorption is expected. However, the hydrodynamic instability severely retards that adsorption. Hence, the collection of macromolecules at the membrane surface is significantly reduced and permeate flux is higher with the use of flow reversal. The remaining parts of the "with-flow-reversal" curves appear to mirror the behavior of the "without-flow-reversal" curves and data from previously published work. This is expected since over the duration of the experiment, protein macromolecules continue to deposit at the membrane surface and some layer compaction occurs due to the pressure driving force.

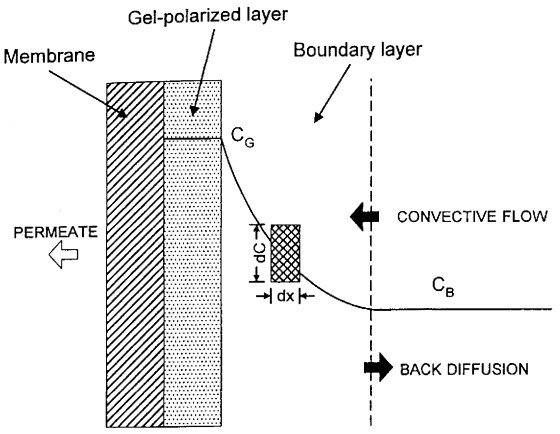

FIGURE 9. Schematic of concentration polarization.

CONCLUSIONS

A comparison of the experimental results of an investigation into the effects of flow reversal on UF operations using BSA solutions has led to the following conclusions:

- Periodic reversal of the direction of flow of the feed stream at the membrane surface results in a significant increase in permeate flux.
- Flow reversal prevents the formation of a stable hydrodynamic boundary layer at the membrane surface.
- The initial rapid drop in permeate flux due to concentration polarization during the first minute of ultrafiltration (Stage 1) is lessened because of the hydrodynamic instability caused by flow reversal.
- The induced hydrodynamic instability slows protein adsorption during the quasi-steady-state period of flux decline (Stage 3).

The results presented here indicate that flow reversal effectively slows the permeate flux decline due to the effects of concentration polarization and membrane fouling. Although these results are solely for UF of BSA, we believe that this technology would be applicable to all membrane separation processes and a wide range of other solutions.

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