

Use of fluid instabilities to enhance membrane performance: a review

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Abstract

This paper presents a review of some of the key methods of generating flow instabilities that have been implemented in membrane separation processes by various workers during the past few years (mainly the 1990s). The types and effects of incorporating these techniques are discussed based on the experimental and theoretical investigations in different applications, particularly, production of potable water from natural fresh water sources, water desalination, and purification of biological and organic solutions and suspensions. The main types of instabilities that are discussed are Dean vortices in helical hollow-fiber membranes, air sparging, novel backflushing techniques and cyclic feed operation. There is a substantial amount of data in the reviewed literature in support of improved performance and fouling prevention in the presence of flow instabilities.

Keywords: Flow instabilities; Flux enhancement; Membrane fouling

1. Introduction

Membrane systems have become well established as essential separation processes that have added new dimensions to the more traditional processes. The recent novel developments in membrane processes have made it possible to achieve many of the design objectives

at economical rates with additional flexibility and improved efficiencies. Examples of successful innovations that have led to outstanding results in the form of large-scale commercial processes can be found in many water treatment applications, particularly brackish and seawater desalination using reverse osmosis membranes and treatment of surface water using micro- and ultrafiltration membranes. In the area of waste water treatment,

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ultra- and microfiltration membranes have already started to gain ground, particularly in the form of membrane bioreactor systems. Many other industrial membrane processes have been developed in fields such as the separation of organic liquid mixtures and purification of biological suspensions.

Among the issues related to membrane processes that have received extensive attention are concentration polarization and membrane fouling. Concentration polarization is influenced by a number of factors such as the flux, trans-membrane pressure (TMP), cross-flow velocity and the flow regime (laminar or turbulent). Higher concentration polarization increases the potential for fouling. Different types of fouling may develop depending on the types of materials accumulating at the membrane wall and the operating conditions. One of the methods to prevent or at least reduce the extent of this behavior is to disturb the onset of the mass transfer boundary layer near the membrane wall. A number of techniques have been used to achieve this goal by introducing various instabilities into the flow. Examples of these techniques are intermittent- and high-frequency backwashing, air sparging, generating secondary flows (Dean and Taylor vortices) and cyclic feed pressure.

The objective of the current study is to review some of the recent literature regarding unsteady operation of the membrane processes. In the following sections, the method of utilizing secondary flows is discussed, followed by the use of methods of gas sparging, backwashing and cyclic (periodic) pressure pulsing.

2. Secondary flows and turbulence promoters

Secondary flows in the form of vortices may occur above certain critical Reynold's numbers for fluid motion in curved channels. Dean vortices are centrifugal instabilities that are

produced in curved channels when the critical Dean number is exceeded while Taylor vortices are generated in the annulus of two concentric rotating cylinders.

2.1. Dean vortices

In a series of experimental and theoretical studies, Belfort and co-workers [1–8] attempted to harness the Dean vortex instabilities to improve the performance of membrane processes. In some of the earlier works, they used a spiral half-tube placed on a flat microfiltration (MF) membrane and were able to show that the flux of Baker's yeast broth and dairy whey increased five-fold compared to cross flow filtration [1]. Chung et al. [4] and Mallubhotla et al. [5,6] confirmed the efficiency of this method with MF membranes by attaching a flat membrane at the edge of the curved end of a U-shaped channel. Flux enhancements of up to 43% were observed using relatively low flow rates. Luque et al. [7] compared the performance of clarifying suspensions of yeast, *E. coli* and mammalian cell cultures in a commercial linear hollow-fiber crossflow MF module with that of a novel helical-wound hollow-fiber membrane module. They reported that the helical hollow-fiber module performed far more superbly with the flux being 3.2-fold higher in the case of constant TMP and 3.9-fold higher in the case of constant flux operation. Kluge et al. [8] also used a helical hollow-fiber MF membrane and studied the effect of varying the viscosity by adding polyethylene glycol to the silica suspensions. They concluded that the flux advantages of the helical design decreased and eventually disappeared as the viscosity of the solution increased to 12 times that of water at 27°C.

In a similar series of works, helical ultra-filtration (UF) membranes were investigated. Manno et al. [9], Guigui et al. [10] and Gehlert et al. [11] showed that the flux in a helical hollow-fiber UF unit increased by a factor of 5 compared

to linear fibers. Moulin et al. [12] found that UF using a helical hollow-fiber geometry increased the flux of bentonite by a factor of 2 and of dextran by a factor of 3 compared to the linear fiber arrangement. Helical nanofiltration modules were also studied by Mallubhotla et al. [13] and found to improve the flux of aqueous solutions. Chung et al. [14] used a three-dimensional numerical model to quantify the concentration polarization for Dean vortex flow in a spiral reverse osmosis (RO) system. They showed theoretically that the presence of Dean vortices promotes mixing and inhibits the growth of the concentration polarization boundary layer.

In a more recent work, Kuakivi et al. [15] employed a new geometry, namely, woven hollow-fiber UF membranes as a means of generating Dean vortices. They found that the woven geometry gave flux increases similar to those of helical fibers but at a lower energy consumption.

2.2. Taylor vortices

Another method that has been used to promote secondary flow is the generation of Taylor vortices in the annulus of two concentric cylinders with a rotating cylindrical filter [16,17]. Flux improvements with biological suspensions similar to those obtained with Dean vortices have been reported. However, the use of rotating elements results in more complicated design requirements and additional capital cost as well as extra power consumption.

2.3. Helical baffles and stamps

A somewhat different method that has also been used to enhance the performance is to promote turbulence using baffles and stamped membranes. Elmaleh and Ghaffof [18,19] used a UF membrane with helical baffles introduced in the filtration element. Suspensions of crude oil and biological cell were treated using this

element. Significant flux improvements were reported. Broussous et al. [20] used a helical stamp on the inside of a tubular ceramic MF membrane. Compared with a smooth-surface membrane, the permeate flux increased by a factor of 6.

3. Air sparging and gas slugs

Gas sparging or injection of air bubbles has been effectively used to reduce concentration polarization and enhance mass transfer. The secondary flows around bubbles promote mixing and reduce the thickness of the concentration polarization boundary layer [21]. When the bubble diameter exceeds that of the membrane (tubular or hollow fiber), then slugs are formed. Large slugs can displace most of the boundary layer and also cause the pressure to pulsate. These effects result in enhancing the flux. Several experimental studies on air sparging have been reported using tubular [21–24], hollow-fiber [25–29] and flat [30] UF membranes.

3.1. Tubular membranes

Li et al. [21] found that the permeate flux of a solution of dextran and human serum albumin in a UF tubular membrane increased with the increase in each bubble size and frequency. Larger bubbles are associated with the generation of vortices, which result in enhanced mass transfer. They noticed that when slugs were formed at larger bubble diameters, further increases in size did not lead to additional enhancements. Cheng et al. [22] studied the flux improvement by gas slugs for dextran solutions in a tubular membrane. They concluded that when the boundary layer resistance is low, e.g., at high liquid speeds, then improvement by gas slugs is limited. However, when the effect of concentration polarization is high, e.g., at low liquid speeds or high TMP, then a significant flux

enhancement is obtained with gas slugs. Ghosh and Cui [23] studied the mechanism of flux enhancement of an upward slug flow in a tubular UF membrane. They developed a theoretical analysis to determine the effect of gas slugs on mass transfer. It was concluded that using gas slugs is more effective at higher TMP but less effective at increasing liquid flow rates. Cui and Wright [24] obtained for a downward flow of feed and gas bubbles inside a tubular UF membrane, flux increases of up to 320%. Again, they concluded that the enhancement was not very significant when the liquid flow was turbulent, i.e., low mass transfer resistance.

3.2. Hollow-fiber membranes

Bellara et al. [25] employed gas sparging with a crossflow hollow-fiber UF of dextran and albumin solutions using different TMPs, feed concentrations and gas-to-liquid flow ratios. Flux enhancements of 20–50% for dextran and 10–60% for albumin were obtained. Laborie et al. [26,27] also used hollow-fiber UF membranes with clay suspensions at a range of air velocities and particle suspensions. Below a critical gas velocity inside the fibers, they found that the flux can be increased by 155%. Above this critical gas velocity, the flux was no longer enhanced. Laborie et al. [28] also performed a theoretical and experimental study inside glass capillaries to explain the hydrodynamics of the gas sparged flow. Serra et al. [29] combined air sparging with backwashing in hollow-fiber modules. They concluded that the backwash time and efficiency were improved when gas sparging was used.

3.3. Flat membranes

Li et al. [30] used gas sparging with flat UF membranes and a mixture of four proteins. They found that gas sparging improved the permeate flux and the efficiency of protein fractionation.

4. Backflushing

Periodic backflushing is widely used in MF and UF processes as a method to clean the membrane. By using a reverse TMP for very short periods of time, the permeate is forced through the membrane in the reverse direction and causes the filter cake or gel to expand, de-clog and eventually be carried away. Periodic or intermittent backflushing is another form of instability that influences the concentration polarization boundary layer, particularly when high-frequency backflushing is used. The efficiency of backflushing is dependent on optimizing a program (e.g., frequency, duration, pressure, single or multiple pulses, uniform or variable pressure and duration) suitable for the specific application. Some of the cases discussed below assist in following the recent trends with this respect.

4.1. Optimized backwashing

Hillis et al. [31] used a programmed backwashing method to optimize the performance of an out-to-in hollow-fiber MF membrane for the treatment of potable water. CFD analysis was used in analyzing the flow. The natural organic matter (NOM) present in the water was treated with coagulation followed by filtration, and the resulting flocs were easily removed by the membrane. In comparison to the normal backwashing in which flow is reversed across the membrane once during each backwash cycle, they arrived at an optimum operation of three consecutive back-washing pulses during each cycle. Kennedy et al. [32] showed that the efficiency of backwashing with a dead-end hollow-fiber UF membrane was significantly improved when it was preceded by crossflushing. Crossflushing was more effective when the flow in the fiber was in turbulent mode. They also found that the efficiency of backwashing was more dependent on the time of the backwash than the pressure. Van Hoof et al. [33] used a dead-end

hollow-fiber MF system with chemically enhanced backwashing (CEB) to optimize the operating conditions and evaluate the suitability of the method as a pretreatment method for seawater RO. Using backwashing intervals and chemicals that are injected during the backwashing period, the SDI was reduced to values lower than 3. Chellam and Jacangelo [34] studied direct-flow out-to-in dead-end hollow-fiber MF. They noticed that when operating at constant flux, a critical velocity existed below which MF fouling rates were small and effectiveness of backwashes was high. Xu et al. [35] developed a theoretical model to optimize the backwash process for a dead-end MF module with suspensions of fine particles. The cake filtration theory was used. Serra et al. [36] used a theoretical model to simulate the performance and operating parameters of an outside-in dead-end UF hollow-fiber module. Chellam et al. [37] developed a theoretical model for a direct flow outside-in hollow-fiber MF system with periodic backwashing. The model was verified using experimental results from a pilot scale unit.

4.2. Air backwashing

Chakravorty and Layson [38] discussed the use of MF as a pretreatment method for RO processes. A system was discussed with a backwash process using compressed air. It is suggested that the rate of filtration can be maintained for long periods with this backwash method. Parameshwaran et al. [39] also used a hollow-fiber MF system with air backwashing. The membrane was submerged in a bioreactor for waste water treatment (membrane bioreactor). The compressed air serves the dual purpose of effectively de-clogging the membrane and aerating the bioreactor.

4.3. High-frequency backpulsing

Ramirez and Davis [40] employed rapid

backpulsing with crossflow MF for removal of suspended solids and dispersed oil from an aqueous stream. Experiments with clay suspensions showed that with rapid backpulsing the permeate flux can be maintained at a value 10 times higher than the long-term flux in the absence of backpulsing. Experiments with dilute oil suspensions showed that rapid backpulsing can increase the permeate flux by up to 25 times, but this enhancement could not be maintained over the life of the membrane. Levesley and Hoare [41] employed high-frequency backflushing for the MF of yeast homogenate suspensions for the recovery of proteins using a ceramic tubular membrane. Short pressure pulses with durations of 0.1 s were applied at a high frequency of 1 Hz to the permeate side by compressed air acting on a diaphragm. The result was an increase of the solute flux by 5.4 times and a slight decrease in the permeate flux. The increase in the transmission of yeast alcohol dehydrogenase was greater than the background protein. Gan et al. [42,43] used a multi-stage high-frequency backwash with a ceramic crossflow MF membrane to treat primary municipal sewage wastewater. A permeate quality was obtained which was better than that required by the EC regulatory standards. Kuberkar et al. [44] used high-frequency short backpulses (0.1–1.0 s) to increase the flux of washed bacterial suspensions and whole bacterial fermentation broth with a crossflow MF system. The flux of washed bacteria improved by 10 times and that of fermentation froth by 2 times compared to a normal crossfiltration operation. They developed a theoretical model to predict the optimum frequency. Redkar and Davis [45] employed high-frequency short backpulsing (0.5–4 s) of crossflow MF using a flat-sheet membrane. The periods of the forward filtration between the backpulses ranged from 1 to 40 s. The flux of washed yeast suspensions was reported to increase by 20 to 30 times compared to the long-term flux without backpulsing. They also

developed a theoretical model to predict and optimize the performance [46]. Mallubhotla and Belfort [47] developed a model for high-frequency backpulsing for cross-flow filtration. The model is based on blocking filtration laws. Parnham and Davis [48] used high-frequency back-pulsing with cross-flow MF to recover protein from bacterial cell debris. The optimum backpulse frequency was 2.5 times per second with a pulse duration of 0.09 s. One-hundred percent protein transmission was achieved compared to only 60% in the absence of backpulsing. Crozes et al. [49] studied the effect of several operating conditions on the irreversible fouling by NOM in hollow-fiber UF membranes, including the frequency of backpulsing, TMP and flux. Short-term reversible NOM fouling was limited by increasing the concentrate velocity, reducing the flux and increasing the backwash frequency. They suggested that the TMP should be kept below a certain level (0.8 to 1 bar for the membrane in their work) to minimize the rate of irreversible fouling.

5. Cyclic operation

5.1. Cross-flow filtration

Hadzismajlovic and Bertram [50,51] used a collapsible-tube pulsation generator upstream of a tubular ceramic crossflow MF unit to enhance the flux of yeast suspension. The crossflow was interrupted periodically (6.3–6.8 Hz) by the pulsation generator. They found flux enhancement of up to 102% for turbulent flow and 450% for laminar flow compared to a steady-flow flux. Gesan-Guiziu et al. [52] used different strategies of cyclic operation on the crossflow MF of skimmed milk using a ceramic tubular membrane for the separation of casein micelles from soluble proteins. A critical ratio of permeation flux over efficient wall shear stress was obtained below which the performance was satisfactory. Above

this ratio, irreversible cake structure was formed which reduced the permeation rate of the soluble proteins. Farley and White [53] used cyclic stop-start operation as a method of increasing the permeate rate and lowering the concentration polarization. Two-hundred percent improvement in the permeate flux and 50% in energy savings were reported. They developed a theoretical model to optimize the process.

Ritcher et al. [54] employed a cyclic feed flow of dispersions of active carbon in water to improve the permeate flux of a dead-end, hollow-fiber UF process. Air sparging was used to clean the membrane.

Defosse et al. [55] employed a combination of periodic step change in the feed pressure with periods of backflushing to improve the permeate flux in hemofiltration using a hollow-fiber hemofilter. The total flux gain in the dynamic regime was reported to reach up to 60% compared to a steady-state operation. Iritani et al. [56] used periodic operation with hollow-fiber UF of bovine serum albumin solutions. The single pass flow filtration was periodically interrupted. A much higher flux was obtained compared to the continuous process.

A number of other specialized methods of pulsatile flow have been reported. Colman and Mitchell [57] used pulsatile flow in combination with baffles for a flat-sheet UF membrane. They found the permeation rate of a dextran solution to increase by 1.5 to 2 times compared to unbaffled steady flow. Spiazzi et al. [58] designed a rotating disc that allowed a periodic pressure pulse to alternate between five tubular UF modules. Tests with aqueous bentonite suspensions showed a 50% reduction in crossflow velocity with the same power consumption per unit permeate flux as required at steady crossflow filtration. Arroyo and Fonade [59] used periodic intermittent jets generated by a pneumatically controlled valve from the main flow. The jets produced large vortices along a tubular UF

membrane. The flux of a bentonite suspension was reported to increase by more than two times compared to steady operation.

5.2. Reverse osmosis

Compared to the relative abundance of information about unsteady operation of UF and MF systems, a much less published work is available for RO. One reason for this could be that the drive to overcome the reversible and irreversible fouling and flux decline is much stronger for MF and UF due to the quick onset of the filter cake or gel and flux decline. RO membranes, on the other hand, have to handle solutions which have extremely low contents of suspended materials, and the fouling problems are normally long-term issues. One of the few studies done in this area is by Kennedy et al. [60,61] who used oscillating pistons to generate a sinusoidal pressure pulse in a tubular RO membrane. An increase of 70% in the permeate rate of a sucrose solution was reported at a frequency of 50 Hz compared to the steady operation. Al-Bastaki and Abbas [62,63] used square wave pressure pulses with time periods of 5 to 15 min in operating a spiral-wound RO membrane to treat brackish water. With a symmetric square wave at an average pressure of 50 bars, a maximum flux improvement of about 13% was obtained [62]. Using an asymmetric wave and an average pressure of 25 bars, an improvement of 6.5% was achieved.

6. Conclusions

Several novel techniques of generating fluid instabilities for membrane processes have been developed and reported in the literature during the past few years. There is a growing amount of evidence about the success of these methods in improving the performance by increasing the flux and reduced fouling. Among the most notable

methods discussed in this work are utilizing: Dean vortices in curved hollow-fiber membranes, turbulence generation using helical baffles, air sparging, high-frequency backflushing, air backwashing and cyclic feed pressure variation. While the short-term enhancement in the flux and reversible fouling was consistent in many of the works discussed above, the advantages of instabilities to reduce long-term fouling and flux decline still need additional work and verification.

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