

## Influence of two phase flow on cake layer resistance and flux enhancement in spiral wound and submerged flat sheet microfiltration membrane modules

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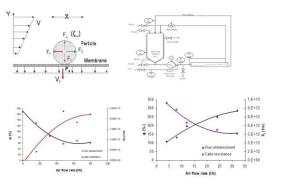
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## **Graphical abstract**



## Abstract

Gas sparging has emerged as an effective technique for control of particle fouling in different microfiltration membrane processes. However most of the research work carried out has been pertinent to hollow fiber and tubular membrane geometries. A little attention has been paid to evaluate the potentails of gas-liquid two-phase for control of particle fouling in spiral wound and submerged flat sheet microfiltration membranes. This study focuses on control of particle fopuling by gas sparging in open channel spiral wound and submerged flat sheet microfiltration membranes. Commercial yeast was used as test suspension. The Effect of gas sparging on membrane fouling and permeate flux was studied by analysing the cake layer characteristics like cake mass deposition, cake layer thickness and cake porosity. The filtration flux and cake properties under various operating conditions, such as cross-flow velocity, filtration pressure, particle concentration, and sparging intensity are analyzed based on hydrodynamics. The results of this study show that gas sparging is very effective in control of particle fouling for both membrane module geometries. It was found that gas bubbling reduced the deposition of particles on the membrane surface due to which cake layer thickness decreases and resultantly permeate flux increased

substantially. The permeate flux increased with increase in gas sparging intensity. A maximum flux enhancement of 170 % and 284 % were observed for spiral wound and flat sheet membranes respectively when gas sparging was applied to the process.

**Keywords:** Microfiltration, gas-liquid two-phase flow, spiral wound, submerged flat sheet, cake layer resistance, flux enhancement.

## 1. Introduction

The application of cross flow MF process is increasing rapidly throughout the world for separation of fine solids from suspension which can not be separated by traditional separation techniques like settling, sedimentation, centrifugation and filtration. Concentration, clarification and purification processes in fruit juice, food, beverages & water processing industries involve suspensions with very fine particles. Cross flow microfiltration and ultrafiltration are most suitable processes for the separation of fine solids from the liquid. Fouling of the membranes induced by particulate deposition, surface adsorption and pore blocking is the major limitation which not only hampers the membrane performance but also reduces the membrane life due to excessive chemical cleaning. There are many proposed techniques which have been found effective in controlling the deposition of particles on membrane surface. These techniques are: turbulence promoters (Finnigan and Howel, 1995), rotating membranes (Kronev et al., 1987), Dean Vortices (Millward et al., 1995) along with unsteady flows, such as pulsating flows (Gupta et al., 1992) and intermittent jets (Arroyo and Fonade, 1993). Although the efficiency of the microfiltration process is improved by the above mentioned techniques, but the industrial application of such solutions is limited by technological aspects. The pore blocking and the adsorption of solids can be only removed either by back-flushing or by application of suitable chemical cleaning agent whereas the external fouling, that is, formation of cake layer on

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membrane surface is influenced by hydrodynamic conditions like flow velocity, applied pressure, particle size and particle concentration etc. The cake layer formed by particle fouling can be removed either mechanically or by chemical treatment. Mechanical cleaning is only possible in the tubular membranes whereas the main drawback of chemical cleaning is that it reduces the membrane life due to aggressive nature of the chemicals against the membrane material (Blanpain-Avet et al., 2009). This has stimulated an increased interest for use of hydrodynamic techniques for control of cake layer formation on the membrane surface thereby reducing the use of chemical cleaning agents in membrane processes. These techniques include back-flushing, pulsatile flow, gas dispersion, etc. The use of gas dispersion in microfiltration process is getting more attention in the present day research for its potential to control the fouling by increasing turbulences on the membrane surface. The use of gas dispersion in hollow fiber was found to be effective in reducing the fouling and enhancing the permeate flux in hollow fibre (Cabassud et al., 1997) and in tubular membranes (Mercier et al., 1997; Vera et al., 2000). As found in the work of M. Mercier-Bonin et al. (1997), high wall shear stresses and low & uniform transmembrane pressure (TMP) have been found to be good hydrodynamic conditions to improve the performances of microfiltration processes. Two phase air-liquid flow generates a slug flow regime in these modules which has been found to be most effective in controlling the fouling. Cui and Wright, 1994 showed up to a 175% increase in permeate flux in yeast microfiltration with gas bubbling. Mercier et al. (1997) applied slug flow in tubular membranes to get a significant increase in the permeate flux. In another study, Mercier et al., 1998 showed 3-time increase in the permeate flux in ultrafiltration of bentonite and yeast suspension by air dispersion in tubular membranes.

It is quite interesting to observe that most of the studies on gas-liquid two-phase flow have been conducted for hollow fiber and tubular membrane modules with feed suspension flowing inside the membrane module (Ndinisa, 2006). To date, more attention has been given to the application of gas-liquid two-phase flow in submerged hollow fiber systems. There is only few published work on application of gas sparging for fouling control in spiral wound and flat sheet membrane geometries. Qaisrani and Samhaber (2008) were the first to report the influence of air dispersion on control of fouling by air dispersion in microfiltration spiral wound membrane module. They used very final colloidal suspension of starch and bacteria for their study and were able to increase the permeate flux up to 60 %. In an unpublished work, Cui et al. (2003) found a flux enhancement of 25 % during their ultrafiltration of dextran through a spiral wound membrane. In case of application of gas dispersion in flat sheet membrane geometry, there is more published work as compared to that of spiral wound membranes. Lee et al. (1993) were the first to apply gas bubbling in flat sheet membranes. They reported that two phase flow enhanced flux for flat sheet ultrafiltration and microfiltration membranes and then later Meircier-Bonin et al. (2000a) also reported an enhancement in flux with air-liquid two phase flow in ceramic flat sheet membrane during microfiltration of baker's yeast suspension. Later on Ducom et al. (2002) observed benefits of injecting gas during nanofiltration of oil/water emulsion by a flat sheet membrane. All these studies were external type applications where two-phase mixture was pumped along the membrane surface. For submerged flat sheet membranes, the only studies reported are for the membrane bioreactors (MBR) application for wastewater treatment (Ndinisa, 2006). Cheng and Lee studied the effect of channel height and membrane inclination on flux enhancement in a flat plate ultrafiltration membrane module in 2007. This study showed show that the permeate flux increased significantly with increasing the gas addition in the narrow channel (2 mm height) no matter with the membrane inclination. In the large channel (10 mm height), the introduction of gas enhanced the permeate flux effectively as the membrane is installed at 90° or 180° inclination Yamanoi and Kageyama evaluated the effect of bubble flow properties between flat sheet membranes on shear stress for optimizing the hydrodynamic parameters of a membrane bioreactor in 2010. They obtained the results by using an apparatus consisting of a visible-channel, simulated-flat sheet MBR, in which the membrane clearance and bubble diameter could be varied. The shear stress on the simulated membrane surface was measured directly. They found that large bubbles with two-dimensional amorphous shapes between the membranes could make the shear stress large in comparison to the case of bubbles smaller than the membrane clearance. Youravong W. et al in 2010 studied the effect of gas sparging on membrane performance during microfiltration of pineapple juice. They applied a ceramic tubular module for this study. They found that a low gas injection factor could increase the permeate flux while higher gas injection factor did not show any benefit on the permeate flux. Gas injection factor of 0.15 gave the best improvement of permeate flux (up to 138%). Increasing gas injection factor tended to reduce reversible fouling but not irreversible fouling. In addition, gas sparging also affected the fouling related to the formation of cake layer onto membrane surface. The density of cake layer increased as the gas injection factor increased.

The spiral wound geometry with open channel spacers generates very high turbulence in the liquid stream and is considered to be effective for control of fouling. The spacers in spiral wound assembly create a feed-channel between facing membrane leaves & promote turbulent flow which reduces fouling phenomena. Osmonics produced first spiral wound element made form Polypropylene in early 80's with open channel spacers with product code 52T-Y which are shown in Figure 1.

The spiral wound membrane geometries are considered to be effective against particle depositions as the shape of the spacers help to generate high shear forces due to high level of turbulences in the feed channels. These membranes are considered to be the workhorse in membrane world (Wagner, 2001) and have been patented until recently in 2005. There is limited information available regarding the control of fouling by gas sparging in spiral wound and flat sheet membrane geometries. According to W.G.J. van der Meer, the contribution of research on application of two-phase flow in spiral wound geometries is only 4.62 %. Therefore, the effectiveness of air-liquid two-phase for control of fouling in spiral wound modules with different types of spacers needs to be studied in depth for finding the effectiveness of this technique in reduction of membrane fouling and to optimize the hydrodynamic parameters like liquid flow rate, gas flow rate, and TMP for enhancement of membrane process. Similarly, it is equally important to investigate the control of particle fouling in submerged flat sheet modules due to their increased applications in Membrane Bioreactors (MBRs) for wastewater treatment. This study therefore focuses on control of particle fouling with the application of gas dispersion in spiral wound and submerged flat sheet membrane geometries. In this study an effort has been made to find out how gas bubbling influences different cake layer properties like cake mass deposition, cake layer thickness and cake porosity. The comparison of performances of two membrane geometries can not be established due to difference in membrane properties and different hydrodynamic conditions.



Figure 1. Two views of the first ever spiral wound element with open channel spacers by Osmonics (now GE Hydraulics and Water Technology, USA).

# 1.1. Mechanism of particle deposition on membrane surface

For devising an appropriate fouling control strategy, it is important to understand the mechanism of particle deposition on membrane surface during filtration process. There are multiple forces which influence the particle motion during the filtration process. Figure 2 shows such forces and their possible direction of action on a single particle in a membrane system.

The adhesive forces which are causing the particle to move towards membrane surface are shown in down and left-ward directed arrows whereas the forces shown with up and right-ward directed arrows are lift forces which are forcing the particle to stay away from the membrane surface. The balance of sum of these forces determines the condition of particle deposition on membrane surface. The particle will deposit on the surface if the sum of adhesion forces is greater than the sum of lift forces otherwise it will keep floating within the suspension. This implies that the deposition of particle on membrane surface can be controlled by enhancing the lift forces in the membrane system. Gas bubbling has the potential to generate high intensity shear forces along the membrane surface which eventually help to minimize the particle deposition on the membrane surface.

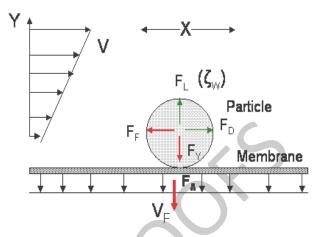


Figure 2. Mechanism of particle fouling and influence various forces on particle.

## 2. Experimental

#### 2.1. Materials

Baker's yeast suspension was used as test suspension. Yeast is composed of almost spherical particles with a mean diameter of  $4.5\mu$ m. Yeast was chosen as a model suspension due to its well-defined granulometric properties. No previous washing of the yeast suspension was carried out in order to evaluate the fouling capacity of both the yeast cells and the extra cellular macromolecules (mainly proteins), which could cause more adhesive cake and severe fouling on the membrane surface.

## 2.1.1. Membrane module

A bench-mounted horizontal spiral wound module and a vertical flat sheet module were used for the experimentation. For spiral wound geometry, PVDF microfiltration elements of type JX 2540 COS from Desalination later named as Osmonics and now, GE Water Technology, USA, with an effective area of 1.01m<sup>2</sup> were used for these tests. The open channel spacer used was of diamond & ladder shape. The pore size of the membrane was  $0.3\mu m$ . The flat sheet module was comprised of 0.016m<sup>2</sup> membrane from Microdyn-Nadir, Germany having a pore size of 0.2  $\mu m.$  The flat sheet module was designed and fabricated locally in the institute. The membranes were cleaned chemically with enzymatic membrane cleaner before start of each experiment and each experiment was conducted at almost same initial pure water flux for both the membrane systems.

#### 2.1.2. Experimental apparatus and method

Figure 3(a) and (b) illustrates the experimental rigs for spiral wound and submerged flat sheet membrane modules respectively for microfiltration of yeast suspension. The temperature of the feed tank was kept constant during all the experiments. Positive displacement pumps (eccentric helical rotor pumps) were used to circulate the feed flow. In spiral wound module, the air was injected at the inlet of membrane module and at outlet of the pump in order to ensure complete dispersion of air in the liquid for generating two-phase flow in both membrane modules. In case of flat sheet membrane, air was injected at the bottom of membrane cell.

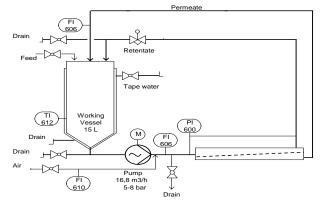


Figure 3(a). Schematic diagram of experimental set up for microfiltration with spiral wound element.

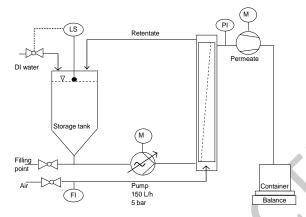


Figure 3(b). Schematic diagram of experimental set up for microfiltration with flat sheet element.

In order to monitor any pressure drop due to air dispersion, a differential pressure measuring pressure gauge was installed at the high-pressure side of the module. For turbidity measurements, WTW-Turb 550 turbidity meter was used. Commercial yeast was used to form the suspension for this study. The average particle size of yeast cell was found to be 4.5µm and Atom Force Microscopy (AFM) was used to determine the size and size distribution of the yeast cells. The experiments were conducted at high feed concentrations of 30 g/L to 40 g/L. The particle size distribution for yeast suspension is shown in the Figure 4. The concentration of solids was calculated as function of turbidity. The suspension system was passed through a 5-µm filter before processing through the membrane. Yeast was chosen as a model suspension due to its well-defined granulometric properties. No previous washing of the yeast suspension was carried out in order to evaluate the fouling capacity of both the yeast cells and the extra cellular macromolecules (mainly proteins) which could cause more adhesive cake and severe fouling on the membrane surface. Pure water flux was measured and recorded before starting the filtration of suspension. Pure water flux provides the reference to assess the effectiveness of the membrane cleaning. All experiments were conducted in recirculation mode. Both permeate and retentate were recirculated in the feed tank while the permeate flow was measured volumetrically.

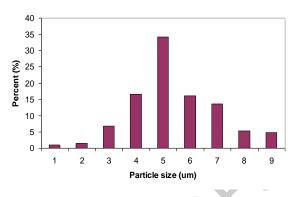


Figure 4. Particle size distribution of yeast suspension obtained from AFM.

It can be deduced from Figure 3 that the mechanism of fouling will be cake formation as the yeast particles are of the average size well above the membrane pore size. Membrane cleaning was performed by using a commercially available enzymatic membrane cleaning detergent Ultraperm-53 by Henkel, Germany. The feed concentration was measured in terms of turbidity units. For this purpose, a WTW-Turb 550 turbidity meter was used to measure the turbidity of feed and the permeate.

## 3. Results and discussion

## 3.1. Effect of feed flow rate on permeate flux

#### 3.1.1. Spiral wound module

Microfiltration experiments with spiral wound element having open channel spacers were conducted to check the effect of feed flow velocity on membrane performance. Figure 5 shows the results of these experiments. It was observed that the permeate flux increased linearly with increase in feed velocity within the range of feed flow rate applied in this study. Figure 5 also shows that the bulk feed concentration of solids increased with an increase in cross flow velocity. The increased cross flow velocity caused increased shear forces along membrane surface deterring the particles to deposit on membrane surface.

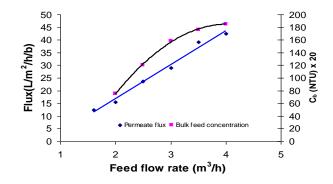


Figure 5. Effect of feed flow rate on the permeate flux in spiral wound membrane;  $\Delta P$ =0.7 bar; C = 40 g/L.

The increased shear forces are due to turbulent flow regime in each capillary for all the feed flow rates applied.

Moreover, the bulk feed concentration in liquid also kept increasing confirming the impact of shear forces on particles along membrane surface. The increase in Reynolds No with flow rate is represented in Figure 6. Therefore, the permeate flux kept increasing with increase in cross flow velocity.

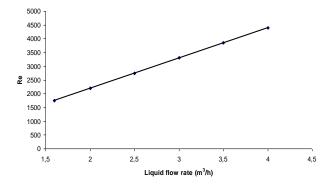
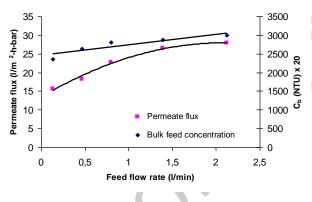


Figure 6. Relationship between Reynolds number and liquid flow rate for spiral wound membrane.

#### 3.1.2. Flat sheet membrane module

In order to see the influence of cross flow velocity on membrane fouling in submerged flat sheet membrane, experiments were conducted at varying cross flow velocities. The typical results of these experiments are shown in Figure 7.



**Figure 7.** Effect of feed flow rate on the permeate flux in submerged flat sheet membrane;  $\Delta P=0.7$  bar; C = 40 g/L.

The impact of cross flow velocity on permeate flux and on bilk feed concentration seems to be similar to that in case of spiral wound element with open channel spacers. It has been proved by many researchers that wall shear force increases as cross flow velocity is increased. Hwang et al. (1996) developed a predictive model for steady state flux in which they correlated the steady state flux to be dependent on wall shear stress rate in terms of cross flow velocity. In this study, they applied moment balance of hydrodynamic and interparticle forces exerted on a single particle for development of their model. This study showed that the permeate flux increased with increasing the cross flow velocity of the liquid for all membrane pore sizes and particle sizes due to increase in wall shear force. Similar results have been observed in Figure 6 that as feed flow rate was increased; it caused an increase in wall shear force due to which deposition of solids on membrane surface decreases. Resultantly membrane performance was improved and permeate flux increased with increase in feed flow rate.

#### 3.2. Effect of gas bubbling on particle fouling

Experiments were conducted to investigate the influence of gas sparging on particle fouling for both membrane modules at varying air flow rates. Flux enhancement factor  $\phi$  and cake resistance were considered as the indices for decrease in fouling. Flux enhancement factor,  $\phi$  is described as:

$$\phi(\%) = \left[ \left( \frac{Flux_{air}}{Flux_{without.air}} \right) - 1 \right] .100$$

Whereas the cake resistance  $R_{\rm c}$  was calculated from Darcy's law as under:

$$J = \frac{\Delta P}{\mu \cdot (R_m + R_c)}$$

Figure 8 shows the effect of air dispersion on flux enhancement and cake resistance for spiral wound membrane whereas Figure 9 shows the influence of air dispersion on cake resistance and permeate flux for submerged flat sheet membrane.

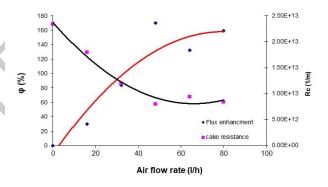


Figure 8. Influence of air dispersion on flux enhancement and cake resistance for spiral wound membrane at QL = 2 m<sup>3</sup>/h;  $\Delta P$  = 0.7 bar; C = 40 g/l.

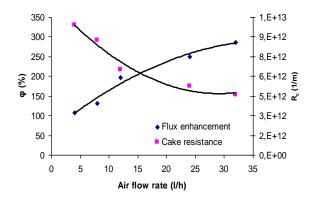


Figure 9. Influence of air dispersion on flux enhancement and cake resistance for submerged flat sheet membrane at  $Q_L$  = Nil;  $\Delta P = 0.7$  bar; C = 40 g/l.

It can be seen from both the figures that injection of air reduced the cake layer deposition on membrane surface resulting a decrease in cake layer resistance for both the membrane geometries which caused an increase in the permeate flux as air flow rate was increased. A maximum flux enhancement of 268% was achieved for flat sheet membrane module at an air flow rate of 32 l/h whereas a maximum flux enhancement of 170% was attained for spiral wound membrane module at an air flow rate of 50 l/h. Although the TMP and feed concentrations were kept same for both modules, however due to varying hydrodynamic conditions due to membrane geometries, the rise in flux can be compared.

It is also observed from Figures 8 and 9 that this enhancement in permeate flux in both membrane modules is due to decrease in cake-layer resistance associated to continuous air bubbling during the filtration process. The air bubbles generate very high flow instabilities and turbulences in the liquid stream which ultimately effect the cake properties like cake-layer thickness, cake deposition on membrane surface and cake-layer porosity in a way that the cake-layer resistance is decreased and membrane performance is enhanced in terms of permeate flux. The influence of air bubbling on cake layer properties and cake-layer resistance will be presented in some other paper.

#### 4. Conclusions

The increase in cross flow velocity significantly reduced cake layer resistance by reducing the quantity of deposited solids on membrane surface for both flat sheet and spiral wound membrane modules. It was observed that the injection of air bubbling improved the filtration process significantly for both membrane modules by reducing the cake-layer resistance. A flux enhancement of 170% and 268% was obtained for spiral wound and flat sheet membrane modules at an air flow rate of 32 l/h & 50 l/h respectively. Air bubbling is more effective in improving membrane performance as compared to increased cross flow velocity method in both membrane geometries. The economic feasibility of air bubbling technique is yet to be established for different membrane modules for same suspension system and with similar membrane effective surface.

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