

## **Cake layer reduction by gas sparging cross flow ultrafiltration of skim latex serum**

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### **Abstract**

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A gas sparged method was investigated for reducing cake layer formation and enhancing the cross-flow ultrafiltration process. The injection of nitrogen gas promotes turbulence and increases the permeate flux of the process fluid. Experiments were carried out using a tubular membrane (100 kDa MWCO), mounted vertically with skim latex serum, which results from the coagulation of skim latex by-product. The objective of this research was focused mainly on the observed reversible cake resistance during the cross flow ultrafiltration of skim latex serum. The effect of operating parameters, including feed flow rate, flowrate gas sparging and transmembrane pressure were investigated. Results obtained thus far show that the use of gas sparged technique has been able to enhance total permeate flux in the range 8.29% to 145.33% compared to non-gas sparged condition. In this research optimum permeate flux was obtained at a feed flowrate of 1400 ml/min, a flowrate gas sparging of 500 ml/min and a transmembrane pressure of 0.89 barg.

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**Key words :** enhance permeate flux, cake layer resistance, gas sparging, fouling reduction

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Fouling is partly due to the formation of a slowly thickening layer on the membrane surface. The extent of membrane fouling depends on the nature of the membrane used and on the properties of the process feed. The first means must be carefully choice of membrane type. Secondly, a module designs which to be provided suitable hydrodynamic conditions for particular application should be chosen. This fouling can occur as accumulation of cells, cell debris, or other rejected particle on the top surface of the membrane (external fouling), or as deposition and adsorption of small particle or macromolecules within the internal pore structure of the membrane (internal fouling). In practice, permeate flux or permeation rate falls with time due to membrane fouling that is blocking of the membrane surface and pores by the particulate materials. In membrane filtration, fouling is a major problem during filtration process. Fouling cannot be prevented from the surface of the membrane, but it can be reduced by certain techniques such as gas sparging. Several studies on the use of gas sparging have been reported to enhance ultrafiltration performance resulting in higher permeate fluxes (Cui and Wright, 1994, 1996). Early studies only concentrate on the use of dilute solution or pure liquids as media. This work focuses on the effect of flowrate gas sparging as a mean to minimize fouling during crossflow ultrafiltration of skim latex serum.

Membrane process can be operated in both cross flow mode, dead-end flow. In dead-end filtration (conventional filtration) the feed flow is perpendicular to the membrane surface which causes build up of debris at the membrane surface effecting a reduction fluid permeation. Due to membranes invariably having small pores, they will plug and blind off instantly.

Crossflow mode of filtration avoids the build-up of solid particles or debris on the membrane surface. Crossflow filtration employs tangential flow across the membrane surface which provides a continuous scouring action and hence reduces the membrane fouling layer due to feed stream debris and macromolecules. Particles de-

posited on the membrane surface are swept away by the feed flow. Crossflow filtration is influenced by a great number of parameters such as crossflow velocity, transmembrane pressure, membrane resistance, cake layer resistance, size distribution of the suspended particles and particle form (Zeman *et al*, 1996, and Cheryan, 1998).

The advantages of cross flow filtration mode over conventional filtration are:

- (a) a higher overall liquid removal rate is achieved by prevention of the formation of an extensive cake layer.
- (b) process feed remains in the form of a mobile slurry suitable for further processing.
- (c) solids content of the product slurry may be varied over a wide range.
- (d) it may be possible to fractionate particles of different size.

Many different models have been proposed to predict permeate flux during ultrafiltration and microfiltration. The cake layer and membrane may be considered as two resistance in series, and the permeate flux is then described by Darcy's law:

$$J_p = \frac{1}{A_m} \frac{dV_p}{dt} = \frac{TMP}{(R_m + R_c)\mu_p} \quad (1)$$

The cake filtration theory has been successful in describing permeate flux decline during ultrafiltration or microfiltration of particulate suspensions. The values of *TMP* and  $R_m$  remain constant during filtration with assumption no resistance of internal fouling. Transmembrane pressure (*TMP*) can be calculated by the following equation:

$$TMP = \frac{P_1 + P_2}{2} - P_3 \quad (2)$$

$R_c$  can be broken down to reversible cake layer resistance ( $R_{rc}$ ) and irreversible cake layer resistance ( $R_{ic}$ ), and is calculated by the following equation:

$$R_c = R_{rc} + R_{ic} \quad (3)$$

$R_{ic}$  is estimated after the membrane has been

fouled, by replacing the feed with water to eliminate reversible cake layer resistance. There is not a direct procedure to estimate irreversible fouling or irreversible cake layer resistance quantitatively. A qualitative picture may be attempted by measuring water flux after through washing of the membrane at the end of an experiment. After cleaning the water flux was checked to ensure that the intrinsic membrane resistance ( $R_m$ ) had recovered to its original value. Therefore, the water permeate flux of the fouled membrane is determined to estimate the irreversible cake layer resistance using the following equation:

$$R_{ic} = \frac{TMP}{\mu_p J_w} - R_m \quad (4)$$

where  $J_w$  is water permeate flux of the fouled membrane and the reversible cake layer resistance can be calculated by using equation (3).

### Materials and Methods

The experimental apparatus is shown schematically in Figure.1. Experiments were carried out using 100 kDa MWCO PVDF a tubular membrane (PCI Membrane System), with an internal diameter of 0.0125 m, length of 1.2 m and a membrane area of 0.0471 m<sup>2</sup>. The tubular

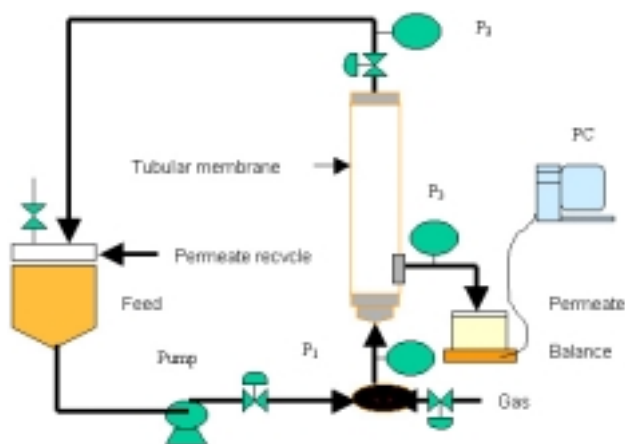


Figure 1. Schematic of pilot scale crossflow ultra-filtration with gas sparging

membrane was installed vertically and was used in all the experiments. All experiments were carried out at room temperature ( $30 \pm 2$  °C) for 4 hours.

Skim latex serum samples derived from coagulation of skim latex was obtained the rubber factory. Due to the turbid nature of skim serum latex, it was necessary to store it over night and pre-filtered before use. Permeate flux was weighed using an electronic balance where the mass was recorded every five seconds by computer and then recycled back to the feed tank to maintain a constant feed concentration.

The range of the physical and operating parameters investigated in this study were:

1. Feed flowrate (ml/min) : 1000, 1200, 1400 and 1600
2. Transmembrane pressure (barg) : 0.51, 0.63, 0.77 and 0.89
3. Sampling time (minutes) : 30, 60, 90, 120, 150, 180, 210 and 240
4. Flowrate gas sparging (ml/min) : 000, 300, 400 and 500

After each experimental run, the membrane was washed with 1.0% NaOH immediately and then flushed with distilled water. After cleaning the flux was checked to ensure that the intrinsic membrane resistance ( $R_m$ ) had recovered to its original value, was calculated by comparing the pure water flux before experiment. Of course, pure water permeate flux before and after experiment is different due to irreversible fouling. It may be noted that there is not a direct procedure to estimate irreversible fouling quantitatively. Irreversible fouling depends on solute-membrane interaction and of course the extent of washing of the membrane at the end of experiment. The irreversible cake layer resistance ( $R_{ic}$ ) was then determined using equation 4. The cleaned membrane was stored in 0.1% sodium metabisulphite solution at room temperature when not in use.

### Results and Discussion

#### Effect of gas sparging on permeate

A typical graph showing the permeate flux

with time and effect of flowrate gas sparging on total permeate flux at feed flowrate of 1400 ml/min and different transmembrane pressure for 240 minutes are given in Figure 2 and Figure 3 respectively.

It can be seen from Figure 3, the permeate flux increased due to gas sparging. The injection of nitrogen gas into the feed stream promotes turbulence in the process fluid and also increases the superficial crossflow velocity. The use of gas

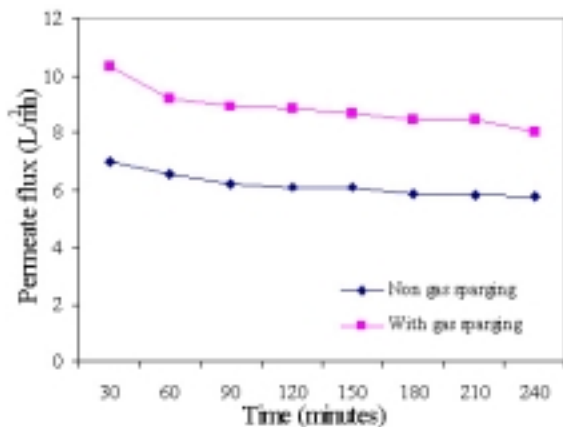


Figure 2. A typical graph showing permeate flux with time at feed flowrate of 1400 ml/min and transmembrane pressure of 0.89 barg.

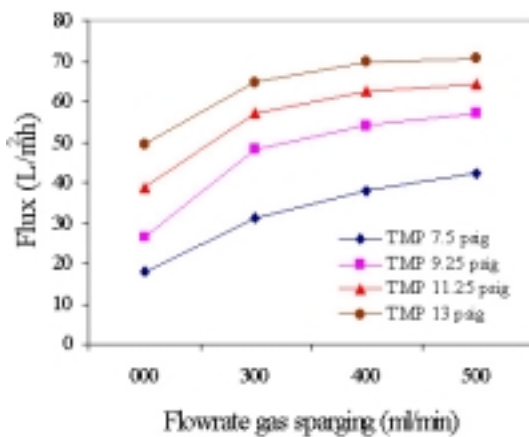


Figure 3. Effect of flowrate gas sparging on total permeate fluxes at feed flowrate of 1400 ml/min and different transmembrane pressure for 240 minutes.

sparged technique to disrupt the cake layer formation, when compared with the non gas sparged condition resulting the total permeate flux increases in the range 8.29% and 145.33%. It can also be seen in Figure 2 that for non-gas sparging and with gas sparging condition permeate flux decreases gradually with time. The decrease in permeate flux with time is due to the formation of cake layer on membrane surface. The cake layer formed on the membrane surface increased as the amount of the retained particles increases with time. The cake layer formed on the membrane surface, referred to as external fouling, is usually reversible. Resulting in permeate flux, for all flowrate gas sparging condition permeate flux continues to decrease with time. So, permeate flux decline caused by membrane fouling cannot be recovered by gas sparging. Gas sparging can only improve performance of crossflow ultrafiltration.

**Effect of transmembrane pressure on permeate**

A typical graph showing the effect of transmembrane pressures on total permeate fluxes for 240 minutes is shown in Figure 4.

The applied transmembrane pressure (TMP) was varied from 0.51 barg to 0.89 barg.

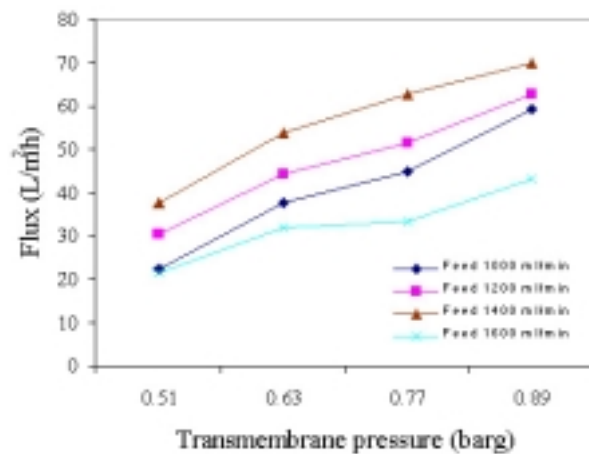


Figure 4. Effect of transmembrane pressure on total permeate fluxes at different feed flowrate and different transmembrane pressure for 240 minutes.

These results show that permeate flux increases linearly with increase in transmembrane pressure. This can be explained by the fact that with higher transmembrane pressure can increase the driving force acting on permeable particles, and eventually permeate flux also increases. From both figures at a feed flow rate of 1400 ml/min and transmembrane pressure of 0.89 barg a resulted in highest flux.

### Cake layer resistance formation ( $R_c$ )

#### a. Effect of gas sparging on cake layer resistance

Cake layer resistance during the cross-flow ultrafiltration of skim latex serum at a constant feed flowrate of 1400 ml/min and transmembrane pressure of 0.89 barg at different flowrate gas sparging is shown in Figure 5.

Generally, it can be seen that cake layer resistance increases gradually with time, signaling the formation and growth of cake layer thickness on the membrane surface.

It has been mentioned previously, permeate flux declined due to the increase in cake layer resistance ( $R_c$ ) attributed to cake formation on the membrane surface. Occurrence of cake layer formation on membrane surface cannot be

avoided, but can be minimized with promotion of turbulence in the feed stream. In Figure 5 it can be seen that an increase in flowrate gas sparging will decrease cake layer resistance or, in other words, the cake layer became thinner, resulting in an increase in permeate flux. In this work, the greatest decrease in cake layer resistance was obtained at gas flowrate of 500 ml/min. During crossflow ultrafiltration of skim latex serum, membrane fouling can be caused by pore size constriction, pore blocking or the deposition of cells, cell debris or other particles present in skim latex serum.

#### b. Effect of transmembrane pressure on cake layer resistance

The effect of transmembrane pressure on cake layer resistance at a constant gas flowrate of 500 ml/min and different feed flowrate is shown in Figure 6. Cake resistance was found to decrease sharply with feed flow rate, probably due to the cake layer growth being arrested because of tangential flow of the fluid. The feed flow rate increases with transmembrane pressure due to increases in turbulence in the feed stream.

A cake layer resistance minimum is obtained at feed flowrate of 1400 ml/min. The figure

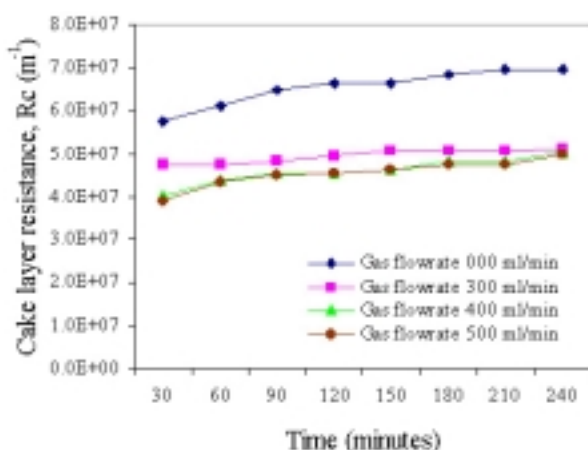


Figure 5. Effect of gas flowrate on cake layer resistance at feed flowrate of 1400 ml/min and transmembrane pressure of 0.89 barg for 240 minutes.

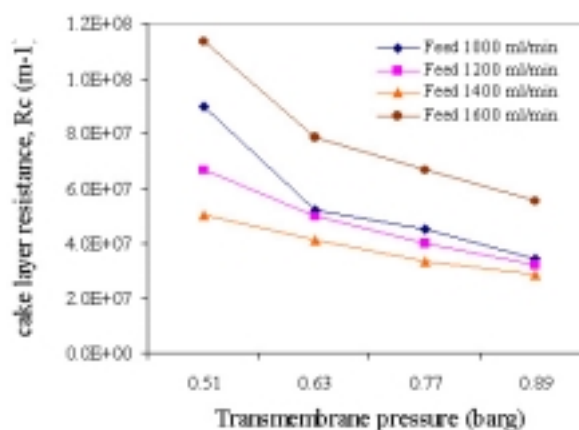


Figure 6. Effect of transmembrane pressure on cake layer resistance at a constant gas sparging and different feed flowrate for 240 minutes.

also shows that cake layer resistance is maximum at feed flow rate of 1600 ml/min. The increase in the cake layer resistance at feed flowrate of 1600 ml/min due to the driving force action on permeable of skim latex serum solution lower than other feed flowrate. Besides that, high feed flowrate and a constant gas flowrate in small tube diameter membrane is unable to promote turbulence in feed stream, unless gas flowrate or gas injection ration is increased. Lower turbulence in the feed stream will result in earlier cake layer formation on the membrane surface, resulting in cake layer resistance decreasing sharply.

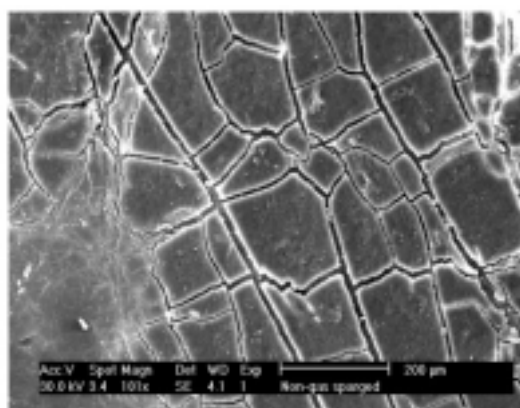
### c. Scanning electron microscope (SEM)

A scanning electron microscope (SEM) type XL40 Philips was used to obtain images of the fouled membrane. From the images shown in Figure 7, formation of the cake layer on the membrane surface after ultrafiltration under non-gas sparged (NGS) condition was more distinct. On the other hand, the cake layer formed on the membrane surface under gas sparged condition was slight and evenly spread. This is probably due to gas sparging not only promoting turbulence but also sweeping away some cake layer or fouling on the membrane surface.

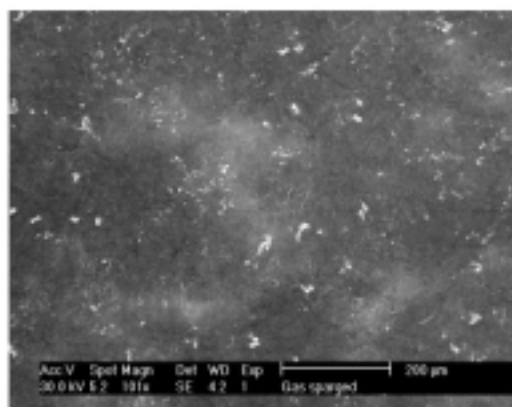
### Conclusions

From the results was obtained in the present study during the cross flow ultrafiltration of skim latex serum several conclusions can be made:

1. The injected nitrogen gas or gas sparging promotes turbulence in the feed stream, suppresses the polarization layer and enhances the permeate flux.
2. Using the nitrogen gas injection or gas sparging technique, the homogeneous liquid phase was changed to heterogeneous gas-liquid phase.
3. The technique of gas-liquid two-phase crossflow ultrafiltration, via the injection of nitrogen gas into the feed stream was found to be effective in enhancing crossflow ultrafiltration and can reduce the occurrence of fouling during filtration.
4. Gas sparging technique when compared with non-sparged condition results in total permeate flux increase of between 8.29% and 145.33%.
5. From this study the recommended optimal conditions for the system are transmembrane pressure of 0.89 barg, feed flowrate of 1400 ml/min and flowrate gas sparging of 500 ml/min.



(a)



(b)

**Figure 7.** SEM photograph of PVDF membrane after filtration. (a) non-gas sparging condition and (b) with gas sparging condition at feed flow rate of 1400 ml/min and transmembrane pressure (TMP) of 0.89 barg.

**Table 1. Characteristics of the skim latex serum after crossflow ultrafiltration with gas sparging and non-gas sparging condition.**

Parameters	With Gas Sparging (WGS)			Non Gas Sparging (NGS)	
	Raw	Treated	Removal (%)	Treated	Removal (%)
pH	3.75	4.46	+15	4.42	+16
Suspended Solids	178	30	83	14	92
Total Solids	7100	370	95	300	96
COD	3998	1332	67	1337	67
BOD <sub>5</sub>	3642	853	77	1012	72
Total Nitrogen	596	295	51	240	60
Ammoniacal Nitrogen	435	115	74	110	75

(All values except pH are expressed in mg/l)

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### List of symbols

- $J$  permeate flux (L/m<sup>2</sup>h)
- $J_w$  is water permeate flux of the fouled membrane
- $A_m$  membrane surface area (m<sup>2</sup>)
- $V_p$  volume of permeate (ml)
- $dt$  filtration time (s)
- $R_m$  intrinsic membrane resistance (m<sup>-1</sup>)
- $R_c$  resistance due to fouling and cake layer formation (m<sup>-1</sup>)
- $R_{rc}$  reversible cake layer resistance (m<sup>-1</sup>)
- $R_{ic}$  irreversible cake layer resistance (m<sup>-1</sup>)
- TMP transmembrane pressure (Pa)
- $P_1$  inlet pressure (barg)
- $P_2$  outlet pressure (barg)
- $P_3$  permeate side pressure (barg)
- $\mu_p$  viscosity of permeate (Pa.s)