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To the Graduate Council:

I am submitting herewith a dissertation written by Wen Chang Liao entitled "Flux enhancements in cross-flow microfiltration." I have examined the final electronic copy of this dissertation for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, with a major in Biosystems Engineering.

Greg J. Hulbert, Major Professor

We have read this dissertation and recommend its acceptance:

Luther R. Wilhelm, Daniel Yoder, Paul Bienkowski

Accepted for the Council:

Carolyn R. Hodges

Vice Provost and Dean of the Graduate School

(Original signatures are on file with official student records.)

To the Graduate Council:

I am submitting herewith a dissertation written by Wen-Chang (Wayne) Liao entitled "Flux Enhancements in Cross-flow Microfiltration". I have examined the final copy of this dissertation for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, with a major in Agricultural and Biosystems Engineering.

Greg J. Hulbert, Major Professor

We have read this dissertation and recommend its acceptance:

Accepted for the Council:

um

Associate Vice Chancellor and Dean of The Graduate School

## FLUX ENHANCEMENTS IN CROSS-FLOW MICROFILTRATION

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A Dissertation

Presented for the

Doctor of Philosophy

Degree

The University of Tennessee, Knoxville

Wen-Chang (Wayne) Liao

August, 1999

AG-VET-MED. Thesis 99b .L56

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## DEDICATION

This dissertation is dedicated to my parents,

Yun-Kun Liao and Mei-Mei Hu Liao, for their truly appreciated support in making my educational opportunities possible. The dedication is also extended to my wife, Ching-Yi, and my son, Jeffrey, for their love and encouragement.

### ACKNOWLEDGMENTS

My sincere thanks are directed to my major professor, Dr. Greg J. Hulbert. I cannot thank him enough for his guidance, expertise, encouragement, patience, and understanding throughout the entire program. Without his help, I would not have been able to finish this work. His support, both as a person and major professor, is more than appreciated. My thanks also extends to my other committee members, Drs. Luther R. Wilhelm, Daniel Yoder, and Paul Bienkowski, for their invaluable advice. I would also like to thank all other faculty members from the Department of Biosystems Engineering and the Department of Food Science and Technology. Their excellent correspondence and friendship made the collaboration between the departments a strong relationship.

I would like to thank my fellow students, Aminta Martinez-Hermosilla and Rahul Seshadri for their support during this project. I would like to thank Ms. Davean Tonkery, Ms. Louise Murr, and Mr. Tommy Burch for their technical assistance. Finally, I would like to express my sincere thanks to Dr. Terry Walker, who is now an assistant professor at Louisiana State University, for his true friendship and assistance in the lab work.

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## ABSTRACT

Two-phase flow microfiltration successfully reduced the fouling problem for several microfiltration processes. Two-phase flow, created by introducing air into the fluid, increased the permeate flux 120%, 45%, and 40% for three different fermented biomass solutions at one hour operating time. For cheese whey microfiltration, the two-phase flow method successfully improved the permeate flux approximately 50% with only 5% air. Without the two-phase flow method, the permeate flux increased 20% when the liquid flow rate was doubled. Intermittent use of air was less effective than continual addition. Operating parameters of two-phase flow microfiltration, such as liquid flow rate and air percentage, were optimized based on permeate flux and energy requirements. The two-phase flow technique saved more energy and processing time than simply increasing the liquid flow rate. An economic analysis was performed to estimate the annual costs for scale-up of a cheese whey microfiltration process.

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## NOMENCLATURE

- Air% air percentage in two-phase flow, %
- A<sub>MM</sub> surface area of each membrane module, m<sup>2</sup>
- C concentration of biomass particles, g/L
- C<sub>B</sub> biomass concentration of bulk solution, g/L
- C<sub>BC</sub> bulk cost of chemical, \$/kg
- C<sub>cc</sub> chemical cost, \$
- C<sub>IP</sub> installation and parts, \$
- C<sub>kw</sub> electricity cost per kilowatt-hour, \$/(kw-h)
- C<sub>Labor</sub> labor cost, \$
- C<sub>M</sub> biomass concentration on the membrane, g/L
- C<sub>ME</sub> cost of monitoring equipments, \$
- C<sub>MM</sub> the initial cost of the membrane modules, \$
- C<sub>MR</sub> cost of membrane replacement, \$
- C<sub>NM</sub> non-membrane costs, \$
- C<sub>P</sub> biomass concentration of permeate flow, g/L
- C<sub>Parts</sub> cost of parts replacement, \$
- C<sub>PM</sub> cost of one membrane module, \$
- C<sub>Pump</sub> cost of pump, \$
- C<sub>TC</sub> total capital costs, \$
- C<sub>TE</sub> total energy costs, \$

- C<sub>TO</sub> total annual operating costs, \$
- C<sub>VP</sub> cost of valves and piping, \$
- D diffusivity, m<sup>2</sup>/s
- D<sub>c</sub> chemical dosage for cleaning membrane, N
- E<sub>Ac</sub> required energy for air compressor, kw-h
- E<sub>CP</sub> required energy for cleaning procedures, kw-h
- E<sub>FS</sub> required energy for pumping feed solution, kw-h
- int integer function
- J permeate flux,  $L/(m^2 \cdot h)$
- J<sub>max</sub> maximum permeate flux, L/(m<sup>2</sup>·h)
- K mass transfer coefficient, L/(m<sup>2</sup>·s)
- M<sub>w</sub> molecular weight of chemical, kg/mole
- N<sub>c</sub> number of cleaning cycles in one year
- N<sub>Labor</sub> number of laborers
- N<sub>MM</sub> number of required membrane modules
- P<sub>1</sub> pressure at position 1, kPa
- P<sub>2</sub> pressure at position 2, kPa
- P<sub>air</sub> air pressure, kPa
- Pe Peclet number
- P<sub>inlet</sub> inlet pressure, kPa
- Poutlet pressure, kPa
- P<sub>permeate</sub> permeate pressure, kPa

- P<sub>TMP</sub> transmembrane pressure, kPa
- P<sub>TPF</sub> two-phase flow pressure, kPa
- $\Delta P$  pressure change, kPa
- Q volume flow rate, L/s, ml/min
- Q<sub>o</sub> original liquid flow rate, ml/min
- Q<sub>req</sub> volumetric treatment rate (plant capacity), m<sup>3</sup>/h
- $\Delta Q$  flow rate change, ml/min, L/s
- R<sub>F</sub> fouling resistance, kPa(m<sup>2</sup>·s)/L
- R<sub>Fo</sub> original fouling resistance, kPa(m<sup>2</sup>·s)/L
- R<sub>s</sub> irreversible system resistance, kPa(m<sup>2</sup>·s)/L
- Re Reynolds number =  $\frac{d_h u \rho}{\mu}$ where u is velocity, m/s, d<sub>h</sub> is hydraulic diameter, m, and  $\mu$  is viscosity, (N·s)/m<sup>2</sup>

Sh Sherwood number = 
$$\frac{K d_h}{D}$$

T<sub>F</sub> filtration time, h

- T<sub>Life</sub> membrane life time, yr
- TS% total solids content, %

V<sub>air</sub> air flow rate, L/min

V<sub>c</sub> amount of cleaning fluid, m<sup>3</sup>

V<sub>Liquid</sub> liquid flow rate, L/min

- V<sub>TPF</sub> air flow rate in two-phase flow, L/min
- <V> average velocity, m/s
- W<sub>Labor</sub> annual labor wage, \$
- $\alpha$  dependence of flux on velocity, expressed in terms of  $\alpha$  (J $\approx$ Re<sup> $\alpha$ </sup>)
- β flow effectiveness constant, min/ml
- ρ density, kg/m<sup>3</sup>
- δ cake thickness, m
- $\eta_1, \eta_2$  pump efficiency

## CHAPTER 1

## INTRODUCTION

Cross-flow microfiltration is a pressure-driven separation process. Unlike conventional "dead-end" filtration, cross-flow microfiltration utilizes a high fluid circulation rate tangential to the filtration barrier to minimize the accumulation of particles at the filter surface. Cross-flow microfiltration potentially enables the continuous separation of solids from suspensions, at conditions that approach a steady-state. The key parameters for cross-flow microfiltration include membrane structure, physical properties of the solutions, transmembrane pressure, liquid flow rate, and temperature. Optimizing the operating parameters is necessary to assure a highly efficient microfiltration process.

Because it is difficult to separate particles smaller than 10 µm by conventional filtration techniques, the use of cross-flow microfiltration has been of considerable interest for several years. Cross-flow microfiltration has been developed to separate fine particles from relatively dilute solutions and is widely used in wastewater treatment. Recently, cross-flow microfiltration has been successfully used in several applications to purify, concentrate, or recover food products. In addition, cross-flow microfiltration has many potential uses for concentration or separation in pharmaceutical and biological processing applications.

Although cross-flow microfiltration is a well-established technology, many

limitations continue to exist. Similar to most conventional filters, a major problem of the cross-flow filtration process is fouling due to formation of a cake-layer of solids at the membrane, which results in a concentration gradient and reduces the permeate flux. The effect of this concentration gradient is called concentration polarization, and it is the major cause of the permeate flux decline. Various techniques have been used to reduce fouling and enhance the performance of cross-flow microfiltration. These techniques include fluid and/or pressure pulsation, back-flushing, and addition of baffles to the membrane. In addition, high liquid velocity also tends to prevent fouling and aids in the cleaning process. However, more energy is required to achieve the high liquid flow rate.

Two-phase flow, which is achieved by injecting air bubbles into the liquid flow, has been identified as an effective technique to reduce the fouling problem (Cui, 1993; Cui and Wright, 1994, 1996; Bellara et al., 1996). Air bubbles not only change the flow pattern without placing baffles inside the membrane, but also provide pulsation which will generate extra shear force to shear the cake. Experimental results for filtration of dextran solutions showed that air bubbles could disturb the concentration polarization layer and significantly increase the permeate flux (Cui, 1993; Cui and Wright, 1994, 1996). They concluded that greater permeate flux enhancements resulted at higher transmembrane pressure and higher feed concentration. The advantages of using two-phase flow microfiltration instead of increasing the pump output are a lower energy requirement and increased pulsation. Two-phase flow microfiltration is a continuous process and the

energy requirement is lower than that required to increase the liquid flow rate. Disadvantages associated with two-phase flow microfiltration are the complexity of system design and foaming problems, though modification of the process and some chemical pretreatments can minimize the disadvantages. To perform an effective two-phase cross-flow microfiltration, optimization of the operating parameters is required.

Two types of solutions, fungal biomass solutions and cheese whey solutions, were used in this research. Filamentous fungi have the potential to produce large amounts of  $\omega$ -3 fatty acids such as eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA), which have several beneficial health effects. Microfiltration can be used to concentrate the fungal biomass solutions to allow subsequent extraction of valuable fatty acid components. Cheese whey, which contains a very diverse mixture of organic (vitamins and proteins) and inorganic compounds (minerals), is the portion of milk remaining after coagulation and removal of curd. Due to the high economic value of proteins, protein recovery from cheese whey has drawn a lot of attention. Microfiltration can be used as a physical pretreatment to eliminate fat from cheese whey and allow subsequent ultrafiltration for protein concentration.

The cost analysis of the cross-flow microfiltration includes capital and operating costs. Capital costs represent the investment required to install a crossflow microfiltration unit, and operating costs represent the expenses associated with energy consumption, labor, chemicals, and maintenance.

### 1.1 Objectives

The main objective of this research was to develop a two-phase cross-flow microfiltration system to concentrate fungal biomass solutions and pretreat cheese whey solutions. Previous work has been exclusively with ideal food systems. The effects of two-phase flow on cross-flow microfiltration of real industrial products was determined. Based on energy and processing requirements, the operating parameters, such as transmembrane pressure, liquid flow rate, and air flow rate, were optimized. The secondary objective was to perform an economic analysis for scaling-up a cross-flow microfiltration system.

### CHAPTER 2

## BACKGROUND

The major problem in cross-flow microfiltration is fouling. In order to reduce or prevent the fouling problem, a two-phase flow microfiltration method was proposed. The performance of two-phase flow microfiltration was evaluated by using fungal biomass and cheese whey solutions, and the economic aspects were also analyzed to determine the benefit of two-phase flow microfiltration. In addition, the basics of membrane technology, applications, influence of operating parameters, and other anti-fouling techniques were studied as well.

### 2.1 An overview of membrane technology

Membrane filtration, which is a simple mechanical process without any additive, is a widely used solid-liquid separation process. Membranes serve as a molecular sieve to separate solute molecules of different molecular size. There is an increasing awareness that membrane systems can provide a useful concentrate and a valuable filtrate or permeate at the same time (Houldsworth, 1994).

When pores of the membranes are much larger than the molecular size of the permeating substance, the membranes are referred to as 'porous'. Hydraulic flow of solvent and low molecular weight solutes occurs through these pores, whereas solutes of high molecular weight cannot pass (Karel et al., 1975). Based on the pore size, membrane separation is generally divided into four categories:

microfiltration (MF), ultrafiltration (UF), nanofiltration (NF), and reverse osmosis (RO) (Houldsworth, 1994). Microfiltration (MF) retains particles in the range from 0.05 to 8µm, including bacteria, yeast and molds, but allows permeation of salts, sugars and smaller proteins. Operating pressures are very low, typically from 30 to 500 kPa depending on application. Ultrafiltration (UF) retains suspended particles, bacteria and the larger molecular weight materials, but allows permeation of salts, sugars and material with a molecular weight less than the membrane pore size. Operating pressures range from 200 to 1000 kPa. Nanofiltration (NF) retains many chemicals, but allows through a proportion of the low molecular weight materials present and water. Operating pressures range from 1000 to 3000 kPa. Reverse Osmosis (RO) retains most chemical species but allows water to pass through, and operating pressures range from 1500 to 6000 kPa. Fig. 1 shows the retained particle size for these four membranes. Both microfiltration and ultrafiltration are widely used in pharmaceutical, chemical, and food industries for the separation of vaccines, fermentation products, enzymes, and other proteins (Shuler and Kargi, 1992). Microfiltration is an especially energy-efficient, economical separation method used to concentrate chemical and biological materials with a high degree of purity.

Membranes can be made from ceramics, stainless steel, or more commonly from polymers like PVDF, PTFE, nylon, polypropylene, polysulfone, and polyethersulfone. Polymers are less robust but are cheaper than inorganic membranes, and their surface chemistry can be modified to make them attract or



Figure 1. Membrane classification based on membrane pore size (Cheryan, 1998).

repel certain species (Butcher, 1990). Recently, ceramic membranes with high chemical and temperature resistances have been introduced. Ceramic membranes are stronger than polymer membranes and can withstand a higher operating pressure. The most common membranes used in food industry are made from polysufone or ceramics. Membranes have become much tougher in recent years. They are able to accept harsher cleaning chemicals (including caustic or acid washes), withstand higher temperatures (some up to 150°C) and steam sterilization is now a feasible operation with some types of membranes.

Over the past ten years in particular, membrane filtration has become more of a standard unit process, and current trends are expected to continue along the following lines:

- Membranes will replace a number of conventional processes in those areas where a technical or economic benefit is demonstrated, such as certain applications involving evaporation, centrifugation, or ion exchange.
- The present proven membrane applications base will continue to expand as the benefits are more widely appreciated and realized.
- New processes, which would not have been possible without membranes, will continue to be developed and offer a commercial edge to those employing them.
- As the range of membrane materials and their properties extends, the possible application areas and the economic benefits will expand at an increasing rate.

Cleaning and sanitizing is an important procedure to ensure the membrane is free from visible impurities, foreign matters, and microorganisms. Theoretically, the permeate flux can be brought back to the previous value after cleaning, but actually, it may not be possible to obtain the initial flux. Several factors, like membrane properties, flow conditions, cleaning reagents, and cleaning time, are important for membrane cleaning (Tragardh, 1989). Chlorine is a universal disinfectant, and is a very effective membrane cleaner. 50 ppm chlorine is required for reducing microbial concentrations down to acceptable levels. For the removal of fats and oils, caustic solutions (like sodium hydroxide) can ensure the most effective and rapid cleaning. Depending on the fouling materials, specific cleaning reagents have to be selected (Cheryan, 1998). The frequency of cleaning is also a critical economic factor because the filtration process has to be stopped for cleaning. In some cases, it may be better to take time off for cleaning and restoring the flux, rather than continuing with a fouled membrane with a low flux.

#### 2.2 Cross-flow microfiltration

Conventional filtration processes operate with the slurry flow "dead-end" into the filtration media. Particles are allowed to accumulate on or in the filtration barrier. It is particularly difficult to use conventional filtration to filter suspensions of colloidal or even micron-sized particles. In addition, conventional filtration usually requires filter aids, which contaminate the solid product. Although conventional filtration techniques can remove particles down to 0.1 µm or less, they are only suitable for treating feeds containing very low concentrations of particles (Bertera et al., 1984). Filtration media which will retain these particles are often very susceptible to plugging. Many of the difficulties associated with conventional filtration can be eliminated if the slurry flow is tangential to rather than perpendicular to the filtration media. With the introduction of cross-flow microfiltration systems, the cake formation can be reduced by using a high velocity transverse flow (cross-flow), and it is capable of concentrating even submicron particles. Cross-flow microfiltration is a pressure-driven separation process. Unlike conventional "dead-end" filtration, cross-flow filtration utilizes a high fluid circulation rate tangential to the filtration barrier to minimize the accumulation of particles at the filter surface. A schematic diagram of a conventional "dead-end" filter and cross-flow filter is shown in Fig. 2.

Cross-flow filtration offers the following advantages (Mackay and Salusbury, 1988):

- Potentially 100% recovery of solids.
- Minimal biological containment.
- Batch or continuous operation.
- Simple temperature control.
- Capacity can be increased by adding further modules.

#### 2.2.1 Applications of membrane separation technology

The use of membrane technology in water treatment applications has been



Figure 2. Comparison of conventional and cross-flow filtration.

of considerable interest in the United States for several years (Yoo, et. al., 1995). Cross-flow microfiltration has been applied to both wastewater treatment (Hart et al., 1988; Al-Malack and Anderson, 1997; Vera et al.,1998) and drinking water treatment (Ma et al., 1998). Metal ions and heavy metals, which are often referred to as "toxic" substances, can be removed by cross-flow microfiltration (Brady et al., 1994; Chang and Hwang, 1996). Also, some microorganisms can be removed by the membrane (Herath et al., 1998; Ghayeni et al., 1999). Cross-flow microfiltration is now an established unit operation for the purification and /or concentration of many liquid food systems, and is also widely used in industry for the clarification and sterilization of liquids. It is particularly useful if the particulate material in the suspension must be recovered as a product.

Both microfiltration and ultrafiltration have been widely used in fruit juice manufacturing process, including apple juice (Wu et al., 1990; Padilla-Zakour and McLellan, 1993; Su et al., 1993), orange juice (Todisco et al., 1996) and pineapple juice (Jaeger-de-Carvalho et al., 1998). The effects of microfiltration and ultrafiltration on the apple juice quality have been studied by several groups (Padilla and McLellan, 1989; Constenla and Lozano, 1995). In addition, the fouling mechanism of apple juice has been studied (Riedl et al., 1998). Cross-flow microfiltration also has been used to recover valuable components like flavor compounds, from fruit purees (Shomer and Merin, 1984; Kawakatsu et al., 1995; Olle et al., 1997).

Microfiltration and ultrafiltration are also widely used in the dairy industry to

manufacture and/or purify dairy products (Bird, 1996; Beuvier et al., 1997; Famelart et al., 1998; Rodriguez et al., 1999). The most common applications are the fractionation of cheese whey and, to a lesser extent, the preconcentration of milk for cheese manufacture (Cheryan, 1998). The efficiency of these membrane processes for producing dairy products has been the subject of several studies in the last ten years (Kosikowski and Mistry, 1990; Le and Daufin, 1996; Guerra et al., 1997; Samuelsson et al., 1997).

Micromembrane technology can also be applied in pulp and paper industry (Sierka and Kommineni, 1998), biotechnology applications (Bowen and Hall, 1995), and alcoholic beverage manufacturing (Müller, 1992).

### 2.2.2 Factors affecting the performance of membrane

Changes in the membrane structure, concentration polarization, and membrane fouling are the three limiting factors which cause permeate flux decline (Mackay and Salusbury, 1988). Membranes have a limited life-time and will wear out with time. The membrane structure, like pore size and supporting materials, can be damaged with inappropriate pressure. In addition, the cleaning procedures will also affect the membrane structure. Most of the changes in the membrane structure are irreversible; therefore, membranes need to be periodically replaced.

The major limiting factor in cross-flow microfiltration is membrane fouling. Fouling is generally attributed to the accumulation of particles on the membrane surface. Flux drops with operating time, usually rapidly in the initial stages and slowly reaches a saturated (steady-state) stage. Fouling is characterized by an irreversible flux decline. Changes in fluid management techniques can alleviate this problem (Kuo and Cheryan, 1983).

Another factor limiting flux is the concentration polarization. When pressure is applied to the fluid stream, solutes are brought to the membrane by hydraulic transport, thus causing a localized increase in solute concentration at the membrane surface. This results in a lower flux either due to increased hydrodynamic resistance or to higher local osmotic pressure decreasing the driving force. This concentration polarization layer is considered dynamic or reversible, and proper fluid management techniques are necessary to enhance mass transfer and minimize the effects of concentration polarization (Cheryan, 1977). The effect of concentration polarization on cross-flow microfiltration has been studied by Gekas and Hallström (1987) and Rautenbach and Albrecht (1989).

Several steps can be taken to minimize these problems (Mackay and Salusbury, 1988).

If the permeate flux decline is due to membrane structure changes:

- Appropriate preconditioning of the membrane.
- Modification the cleaning procedures (use caustic or acid).
- Replacing the membrane

If the permeate flux decline is due to concentration polarization:

- Reducing the solids concentration of the feed.
- Reducing the particle concentration at the membrane surface.

Increasing the mass transfer coefficient.

If the permeate flux decline is due to membrane fouling:

- Selecting a suitable membrane.
- Modification of the membrane surface.
- Removal of particles at the membrane surface.
- Use of optimum hydrodynamic conditions.
- Chemical pretreatment of the feed.

Industrial applications using cross-flow microfiltration are quite difficult to analyze and control. Some efforts have been made in the past five years to establish an automatic control system for cross-flow microfiltration processes (Decloux et al., 1994). Neural networks, which are increasingly used in many engineering applications, have been introduced to study the dynamics of cross-flow microfiltration (Piron et al., 1997). Recently, fuzzy logic has also been introduced to control the cross-flow microfiltration process. Perrot et al. (1996) developed a fuzzy controller, which allows a simultaneous gradual action on the transmembrane pressure and cross-flow velocity, to maintain a constant permeate flux in raw cane sugar syrup cross-flow microfiltration. More control techniques will be available in the future.

#### 2.3 Anti-fouling techniques

Various techniques have been used to reduce fouling and enhance the performance of cross-flow microfiltration. Depending on the factors causing flux decline in specific applications, the anti-fouling techniques can be classified into three categories: modification of membrane, modification of feed solution, and modification of fluid dynamics (Li et al., 1998). These techniques include fluid and/or pressure pulsation, back-flushing, and addition of baffles to the membrane (Milisic and Bersillon, 1986; Rodgers and Sparks, 1992; Bertram et al., 1993; Miller et al., 1993; Park et al., 1994; Davis and Redkar, 1995; Parnham and Davis, 1996; Chellam and Jacangelo, 1998). Some other anti-fouling techniques, including an electrical enhancement method (Okada et al., 1997; Akay and Wakeman, 1997) and a prefilter method (Kwon et al., 1997), have also been developed.

Among these anti-fouling approaches, modification of the fluid dynamics has been the most popular (Li et al., 1998). Inducing pulsatile flow is one of the most promising techniques for modifying the fluid dynamics. The pulsation provides enough shear force to drive the particles off the membrane. Many successful examples have been reported (Jaffrin, 1989; Boonth, et. al., 1991; Rodgers and Sparks, 1992; Gupta et al., 1993; Bertram et al., 1993; Ding et al., 1993). A transmembrane pressure pulsation (Rodgers and Sparks, 1992) and a suspension flow pulsation (Bertram et al., 1993; Hadzismajlovic and Bertram, 1998) have been successfully developed to reduce the fouling problem. Rodgers and Sparks (1992) reported that transmembrane pressure pulsing not only reduced the membrane fouling resistance, but also reduced the concentration polarization resistance. The increase in flux due to pulsing was significantly greater than that due to increasing the flow rate. Bertram et al. (1993) used an unsteady pulsatile flow, which was
generated by using a collapsible-tube pulsation generator, to increase the efficiency of cross-flow microfiltration by 60%. However, the efficiency of these pulsation techniques is decreased with high solids concentration (Bertram et al., 1993). Pulsatile flow microfiltration has also been investigated by Wu et al. (1993) to recover proteins from yeast cell debris suspensions. Li et al. (1998) reported that not all pulsatile flows are equally beneficial to the filtration process. When there is no back-flushing, shear alone will only improve flux performance under certain conditions. In addition, pulsatile flows with briefly interrupted flow rate or pressure waveforms were not so effective in reducing cake resistance.

Back-flushing techniques have been developed into a high-frequency reverse filtration system by Davis and Redkar (1995), who back-flushed the membrane periodically and got a higher permeate flux. Back-flushing techniques have also been used to improve the efficiency of a microfiltration process in a water treatment plant (Vigneswaran et al., 1996). Based on the established back-flushing techniques, a rapid back-pulsing cross-flow microfiltration was introduced by Parnham and Davis (1996) to recover proteins from bacterial cell debris. Ramirez and Davis (1998) used the back-pulsing method to enhance the removal of suspended solids and dispersed oil from an aqueous stream. Very short backpulses (0.1-1.0 sec) have been tested to successfully increase the flux of microfiltration (Kuberkar et al. 1998). In both back-flushing and back-pulsing processes, the transmembrane pressure was periodically reversed, with the purpose of removing particles from the surface of the membrane. Although both

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back-washing and pulsation techniques can be utilized to reduce cake on the membrane surface, back-washing techniques achieve a higher cake reduction (Li et al., 1998). However, back-washing techniques usually require more energy and require periodic interruption of the filtration process.

Turbulent flow can provide extra shear force between the liquid and the particles to reduce cake formation. Thus, the permeate flux decline can be reduced by promoting turbulent flow in the filtration system. Adding baffles to the membrane can change the flow pattern to become turbulent. Instead of placing baffles inside the membrane, turbulent flow can also be created by increasing the liquid flow velocity. High liquid velocity tends to prevent fouling and aids in the cleaning process (Cohen, 1990; Porter, 1990; Li et al., 1998). The drawback is that more energy is required to achieve the high liquid flow velocity.

#### 2.4 Two-phase cross-flow microfiltration

Two-phase flow, which is achieved by injecting air bubbles into the liquid flow, has been identified as an effective technique to reduce the fouling problem (Cui, 1993; Cui and Wright, 1994; Cui and Wright, 1996; Bellara et al., 1996). Air bubbles not only change the flow pattern without placing baffles inside the membrane, but also provide pulsation which will generate extra shear force to shear the cake. The flow pattern of two-phase flow cannot be determined by the usual methods due to pulsation (Butterworth and Hewitt, 1977). However, turbulence can be expected and observed (Fig. 3). The addition of air bubbles to the liquid stream



Figure 3. Flow pattern diagram of two-phase flow in cross-flow filter.

increases the turbulence at the membrane surface as well as the flow velocity. This two-phase flow technique successfully prevents the cake formation and leads to an enhancement of the filtration process.

The creation of a gas-liquid two-phase flow at the membrane surface by air sparging has been shown to reduce concentration polarization and fouling in both microfiltration and ultrafiltration. Air slugs were used to improve the cross-flow filtration of bacterial suspensions (Lee et al., 1993). A flat sheet membrane system and a number of membranes with different molecular weight cutoffs (MWCO) were shown to improve permeate flux by up to 100% with an ultrafiltration membrane (MWCO 300 kDa), and 30% with a 0.2 µm pore size microfiltration membrane. Cui (1993) has shown up to a 250% improvement in flux when compared to conventional cross-flow operation with dextran solutions. Bellara et al. (1996) reported that the permeate flux was enhanced by 20-50% for dextran solutions and 10-60% for albumin. These researchers concluded that greater permeate flux enhancements resulting from injection of air bubbles occurred at higher transmembrane pressure, higher feed concentration, and/or lower liquid velocity. Even at very low air flow rates, significant increases in permeate flux can be achieved with this simple and economical two-phase flow technique.

The effect of gas-liquid two-phase flow on concentration polarization has been studied by Parvatiyar and Govind (1995). They established a mathematical model based on a single particle motion equation derived by Maxey and Riley (1983) to predict the concentration polarization. Their model expressed the concentration polarization as a function of Sherwood (Sh) and Peclet (Pe) numbers.

# 2.5 Advantages and disadvantages of using two-phase flow microfiltration

Two-phase flow microfiltration can be widely applied in the food industry and may have an immediate impact. The advantages of using two-phase flow microfiltration include:

- Providing pulsation and extra shear force to prevent fouling.
- Instead of using air, some other gases can also be used. For instance, it is
  possible to combine the filtration and carbonation processing units by mixing
  carbon dioxide into the liquid stream.
- Continuous process.
- Reducing chemical filtration aids.
- Reducing energy requirement.

Although two-phase flow microfiltration has the above advantages, the application still has some limitations. The disadvantages of using two-phase flow microfiltration include:

- The entire process becomes more complicated.
- It is necessary to find an air outlet in the process line to release the air bubbles. It cannot be a closed system.
- Foaming problems.

The foaming problems can be reduced by adding some food-grade antifoaming chemicals, or by adjusting pH or viscosity. In addition, connecting a foam collector to the processing line may be an alternative. Based on the economical analysis, the cost of designing a two-phase cross-flow microfiltration system can be compensated for energy savings.

## 2.6 Model development

Several mathematical models are available to describe the mechanism of transport through micromembranes (Hunt et al., 1987; Asaadi and White, 1992; Seo, 1992; Henriksen and Hassager, 1993; Prádanos et al, 1995; Song, 1998). Furthermore, some models can be used to simulate specific microfiltration applications, like protein microfiltration (Balakrishnan et al., 1993; Nakamura and Matsumoto, 1998) and apple juice microfiltration (Padilla-Zakour and McLellan, 1993). The mass transfer controlled model and the resistance model are the most widely used theories for modeling permeate flux in microfiltration.

## 2.6.1 Mass transfer controlled model

In the mass transfer controlled mechanism, the permeate flux is affected by the particle concentration gradient. Because particles stick on the membrane during the membrane separation process, there is a concentration gradient near the membrane interface. This effect is called concentration polarization and can be considered as a resistance to transport of permeate through a membrane (Schulz and Rippergen, 1989). In the mass transfer controlled region, the flux becomes independent of pressure. As the pressure is increased the cake packs more tightly, and the resistance is increased in proportion to the pressure.

Permeate flow through a membrane can be modeled according to the mass transfer control theory (Belkacem et al., 1995). Based on the mass transfer model, several theories have been developed to predict the flux decline in cross-flow microfiltration (Bitter, 1991). For the solid-liquid membrane separation shown in Fig. 4, a simple mass balance on the membrane can be expressed as:

$$Biomass_{out} = Biomass_{in} - Biomass_{diffusion}$$
(1)

Applying Fick's first law of diffusion and assuming a binary system:

$$J \times C_{P} = J \times C - D \frac{dC}{dr}$$
(2)

The boundary conditions are:

$$r = 0 \qquad C = C_B$$
$$r = \delta \qquad C = C_M$$

where J is the flux (L/m<sup>2</sup>•h), C is the concentration of biomass particles (g/L), C<sub>B</sub> is the biomass concentration of bulk solution (g/L), C<sub>M</sub> is the biomass concentration on the membrane (g/L), C<sub>P</sub> is the biomass concentration of permeate flow (g/L), D is the diffusivity (m<sup>2</sup>/s) and  $\delta$  is the cake thickness (m). Solving equation 2 and then applying the boundary conditions gives:



Figure 4. Schematic diagram of concentration polarization.

$$J = \frac{D}{\delta} \ln \frac{C_{M} - C_{P}}{C_{B} - C_{P}}$$
(3)

The ratio of diffusivity (D) and cake thickness ( $\delta$ ) is the mass transfer coefficient (K). In addition,  $\frac{C_M - C_P}{C_B - C_P}$  is defined as concentration polarization (Rautenbach and Albrecht, 1989). The theory of concentration polarization has been studied by several research groups (Bhattacharyya et al., 1990; Denisov, 1994). Because the pore size of our microfilter is very small (0.1 µm), the biomass concentration of the permeate flow is assumed to be zero ( $C_P = 0$ ). Then equation 3 can be simplified as:

$$J = K \ln \frac{C_M}{C_B}$$
(4)

Under normal operating conditions the system is not at steady state and the cake thickness is increasing with time. Also, if the solution is recycled (more than once through the membrane)  $C_B$  is increasing with time. Both factors will result in a decrease in permeate flux. According to equation 4, the permeate flux is proportional to the mass transfer coefficient. The permeate flux can be increased by increasing the mass transfer coefficient. Mixing air into the biomass solution will increase turbulence and shear force, which will decrease the cake thickness and increase the mass transfer coefficient. Thus, the permeate flux will increase. In order to predict the permeate flux, the mass transfer coefficient was calculated by

several empirical equations in which the mass transfer coefficient is increased exponentially with Reynolds number (Re) (Henry, 1972; Perry and Green, 1984; Cheryan, 1986).

$$K \propto Re^{\alpha}$$
 (5)

where  $\alpha$  is the dependence of flux on velocity.

#### 2.6.2 Resistance model

Regardless of the magnitude of  $\alpha$ , it is clear that the permeate flux can be increased by increasing the fluid velocity. However, this increase cannot occur indefinitely. This is due to the fact that the velocity will affect the variable fouling resistance ( $R_F$ ) caused by cake layer formation, but will not affect the irreversible system resistance ( $R_s$ ) caused by the pores themselves and by pore plugging and inner-pore adsorption. A resistance based model using these two resistance terms allows the effect of transmembrane pressure (the driving force) to be incorporated.

$$J = \frac{\Delta P}{R_F + R_S} \tag{6}$$

where  $\Delta P$  is the transmembrane pressure (kPa),  $R_F$  is the fouling resistance (kPa(m<sup>2</sup>•s)/L), and  $R_s$  is the irreversible system resistance (kPa(m<sup>2</sup>•s)/L). Other resistance models were also developed by several research groups (Belkacem et

al., 1995; Cheryan, 1998; Sierka and Kommineni, 1998).

At low velocity the flux is somewhat independent of pressure because increased pressure causes the cake to pack tighter and increases  $R_F$ . This effect is illustrated schematically in Fig. 5. As the velocity is increased the fouling resistance is reduced and pressure becomes a significant factor. The previously discussed models are valid only for the mass transfer controlled region. The maximum flux occurs at very high liquid velocity where  $R_F$  becomes negligible.

$$J_{max} = \frac{\Delta P}{R_s} \tag{7}$$

Combining equations 6 and 7, the permeate flux can be calculated as:

$$J = \frac{\Delta P}{R_F + \frac{\Delta P}{J_{max}}}$$
(8)

To model the effect of flow velocity on the fouling resistance ( $R_F$ ,  $kPa(m^2 \cdot s)/L$ ), the following empirical equation is proposed (Liao et al., 1997):

$$R_{F} = \frac{R_{Fo}}{1 + \beta \cdot (Q - Q_{o})} = \frac{R_{Fo}}{1 + \beta \cdot \Delta Q}$$
(9)

where  $R_{F_0}$  is the original fouling resistance (kPa(m<sup>2</sup>•s)/L),  $\beta$  is the flow effectiveness constant (min/L),  $Q_0$  is the original liquid flow rate (no air) (L/min), Q is the combined liquid and air flow rate (L/min) and  $\Delta Q$  is the flow rate change (L/min).



Figure 5. The effect of velocity on permeate flux. (Transmembrane pressure:  $P_3 > P_2 > P_1$ )

The  $\beta$  value is a constant with units of inverse volumetric flow. It provides a measure of the flow effectiveness in reducing the fouling resistance. This model is somewhat intuitive and describes the dynamics of the flux and fluid flow relationship for the mass transfer controlled, pressure controlled, and transition flow regions. Previous models have been limited primarily to the mass transfer controlled region.

#### 2.7. Operating parameters for two-phase cross-flow microfiltration

## 2.7.1 Physical properties of solutions

The density, viscosity, and diffusivity are important factors influencing the permeate flux. These factors not only affect the mass transfer coefficient in the cake layer but also affect the fluid mechanics. For instance, the high viscosity of concentrated solutions will result in a very high pressure drop through the filter's flow channel (Russotti et al., 1995). Therefore, increasing the viscosity will increase power consumption, reduce turbulence, and reduce flux. In addition, the particle concentration has a significant influence on the permeate flux in cross-flow microfiltration (McCarthy et al., 1996). Even the particle shape has an effect on the permeate flux (Connell et al., 1999). Generally speaking, flux will decrease with increasing suspension concentration, but some interesting opposite effects have been reported. Cheryan (1998) reported that the flux could either increase or decrease with increasing concentration. Moreover, McCarthy et al. (1996) reported that the flux increase of enhanced cake

removal rate. This enhanced cake removal rate was a result of increased wall shear stress brought by increased suspension viscosity.

#### 2.7.2 Transmembrane pressure

Because cross-flow microfiltration is a pressure-driven separation process, the transmembrane pressure (TMP) becomes a major factor influencing the permeate flux. The transmembrane pressure is defined as:

$$P_{\text{TMP}} = \frac{P_{\text{inlet}} + P_{\text{outlet}}}{2} - P_{\text{permeate}} \tag{10}$$

where  $P_{inlet}$  (kPa) is the inlet pressure which is measured before the membrane;  $P_{outlet}$  (kPa) is the outlet pressure which is measured after the membrane; and  $P_{permeate}$  (kPa) is the permeate pressure which is taken at the permeate flow.

Increasing transmembrane pressure usually increases the permeate flux. At low pressure, the permeate flux is directly proportional to the transmembrane pressure. A plateau pressure is often reached where the flux becomes invariant with further increases in pressure. When the pressure becomes too high, the cake layer will be tightly packed, which will reduce the permeate flux. A well-packed membrane is hard to clean and sometimes the high pressure may damage the membrane. Pillary and Buckley (1992) and Cumming et al. (1999) reported the relation between the cake layer and the transmembrane pressure. In a two-phase flow microfiltration system, the control of transmembrane pressure is very important because air bubbles are very sensitive to pressure change.

# 2.7.3 Liquid flow rate

Liquid flow rate is the average rate at which the process fluid flows parallel to the membrane surface. Increasing liquid flow rate will induce turbulence in the fluid, and provides shear force to clean the membrane. In general, higher liquid flow rate provides higher permeate flux. There is a significant interaction between liquid flow rate and transmembrane pressure on fouling. At low flows, higher flux can be achieved with higher liquid flow rates. At high flows, the flux is limited by the membrane resistance. The flux can be increased by increasing the transmembrane pressure. In the pressure controlled region, the effect of increasing liquid flow rate becomes minimal and the pressure effect becomes dominant. The biggest drawback to increase the liquid flow is the increased energy requirement.

#### 2.7.4 Velocity head effect

Pressure gauges are generally placed before and after the membrane to monitor the transmembrane pressure. However, this should be considered as an apparent pressure. Fig.6 shows the schematic diagram of the cross-flow microfilter. When the liquid is pumped from the feed tube into the membrane, the crosssectional area changes and the liquid velocity changes accordingly. According to the Bernoulli equation, the pressure will be affected by the velocity change (Bird et al., 1960; Geankoplis, 1993). Therefore, a velocity head effect needs to be



Figure 6. Schematic diagram of cross-flow microfiltration set-up.

considered to calculate the actual pressure in the membrane tube. If we assume there is neither mechanical work nor friction loss and the elevation does not change, the Bernoulli equation can be simplified as:

$$\frac{l}{2}\left(\langle \mathbf{v}_{2}\rangle^{2} - \langle \mathbf{v}_{1}\rangle^{2}\right) + \frac{P_{2} - P_{1}}{\rho} = 0$$
(11)

where  $\langle v \rangle$  is the average velocity,  $\rho$  is the density and P is the pressure. From equation 11, the pressure change can be calculated by:

$$\Delta P = P_2 - P_1 = \frac{\rho}{2} \left( \langle \mathbf{v}_1 \rangle^2 - \langle \mathbf{v}_2 \rangle^2 \right)$$
(12)

Equation 12 indicates that reducing the flow velocity will increase the pressure, so the actual pressure can be calculated by knowing the velocity change.

#### 2.7.5 Air flow rate

Two-phase flow is generated by injecting air bubbles into the liquid stream. Air bubbles provide shear force to clean the cake layer at the membrane surface; thus, the permeate flux will be improved. Air flow can be created by an air compressor which provides high pressure compressed air. A small percentage of injected air will significantly affect the performance of two-phase flow microfiltration. To successfully control the two-phase microfiltration process, it is necessary to accurately monitor the amount of injected air. Because air bubbles are very sensitive to pressure, transmembrane pressure has a major influence on determining the air percentage. The volume of air bubbles changes with pressure, and the volume change will affect the air flow rate measurement. In most applications, it is desirable to maintain the air flow rate as a constant.

## 2.7.6 Temperature

The effect of temperature on permeate flux is significant. Higher temperature will lead to higher flux since higher temperature reduces the viscosity of the fluid. The required energy for pumping can also be lowered. In addition, high temperature (> 55°C) can also minimize the microbial growth problems. Thus, it is better to operate the filtration process at the highest temperature allowed. This high temperature filtration process needs to be balanced against the higher energy costs. On the other hand, a higher temperature can induce precipitation of insoluble salts, denaturation of proteins, or gelatinization of starch to foul the membrane (Cheryan, 1998). In the two-phase flow microfiltration system, higher temperatures also affect the density of air bubbles. It is critical to maintain a constant temperature during the two-phase flow microfiltration process. Sometimes it requires some extra effort to keep the temperature constant, because as the fluid is circulated, the temperature of the fluid will gradually increase due to the friction.

#### 2.8 Application #1: Concentration of fungal biomass solutions

Beneficial health effects from consumption of certain fish oils have been

attributed to the presence of the  $\omega$ -3 fatty acids, eicosapentaenoic acid (EPA) and docosahexaenoic acid (DHA). These  $\omega$ -3 fatty acids have been linked to a reduced risk for coronary heart disease, arthritis, inflammation, hypertension, and cancer (Simopoulos, 1989). Declining marine resources and increasing demand for these fatty acids have prompted the search for alternative sources.

Filamentous fungi have the potential to produce large amounts of EPA within the mycelial walls when grown at optimal conditions (Radwan, 1991; Ghandi and Weete, 1991). These fungi are also capable of being genetically engineered to produce large amounts of fungal oil (Shimizu et al., 1988; Yamada et al., 1987; Shinmen et al., 1992; Ratledge, 1993). Fungal bioconversions are well suited for treatment of processing waste from U.S. food industries. Bioconversions of organic waste streams to useful products provide the possibility of reducing waste disposal costs.

Membrane technology, such as microfiltration, can be used to concentrate fungal biomass solutions to allow subsequent extraction of valuable fatty acid components (Conrad and Lee, 1998). Dry fungal biomass can be recovered by applying both centrifuging and freeze drying technologies to completely remove water from fungal biomass concentrates. A supercritical carbon dioxide extraction method has been introduced to successfully extract the fatty acids from the dry fungal biomass (Walker, 1997).

## 2.9 Application #2: Pretreatment of cheese whey

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Whey is the portion of milk remaining after coagulation and removal of curd. There are two principal types: sweet whey and acid whey. Sweet whey is a byproduct from the production of rennet-type hard cheeses like Cheddar and Swiss, and has a pH of approximately 6.1. Acid whey is a byproduct from the production of acid-type cheeses, such as cottage cheese, and has a pH of approximately 4.4 to 4.6 (Igoe, 1989). The dry form of whey is called whey solids. Both whey and whey solids can be used as a source of lactose, milk solids, and whey proteins. In addition, they can also be used as a replacement for milk solids to provide a source of protein, solids, and flavor. For example, both whey and whey solids have been widely used in baked goods, ice cream, dry mixes, and beverages (Igoe, 1989). One of the newest uses involves the fermentation of the lactose in whey to produce alcohol for mixture with gasoline.

The chemical composition of cheese whey has been widely studied. Cheese whey contains a very diverse mixture of organic (vitamins and proteins) and inorganic compounds (minerals). The ability of whey protein concentrates to form foams is probably one of the most studied properties. The extent of the protein interaction with the air-water interface is affected by the ability of the molecule to reach the interface and unfold to form viscous films. This characteristic can be desirable or undesirable in specific food applications. In two-phase flow microfiltration processing, the foaming ability of whey proteins is a negative factor.

Worldwide production of cheese whey is greater than 90 million tons per year, and cheese production is increasing at a rate of approximately 3%/yr (Zall,

1984). Approximately 47% of the whey which is produced is not utilized (Flatt et al., 1988), resulting in significant disposal costs. The problems associated with whey disposal are large because of the large quantities of whey produced, its high BOD value, its relatively low level of solids, and the marginal economic value of these solids without further upgrading (Potter and Hotchkiss, 1995). In recent years several factors have had an influence on the disposal of whey. Membrane technology, including reverse osmosis, ultrafiltration, and microfiltration, have made it economically feasible to remove fats from whey and to separate whey into its lactose and protein components. Tighter antipollution laws have forced cheese manufacturers to adopt alternatives for whey disposal. In addition, modified and blended whey products with unique properties have found new markets. Membrane technology has attracted the attention of cheese and whey producers because the appropriate membrane can simultaneously fractionate, purify, and concentrate whey components to enhance their utilization and reduce the pollution problem.

Due to the high economic value of proteins, protein recovery from cheese whey has drawn a lot of attention (Palmer, 1977). Because the presence of proteins and fats in cheese whey cause fouling, some pretreatment methods (physical or chemical treatments) need to be considered before using ultrafiltration (Karleskind et al., 1995). Instead of using chemicals, microfiltration can be used to separate fat from the cheese whey solutions. Because the pore size of the microfilter is larger than the size of protein molecules, proteins and other minerals will pass through the micromembrane in the permeate. The permeate solution can be further processed by applying ultrafiltration to recover the valuable proteins.

## 2.10 Economic analysis

The costs to install and operate a membrane separation process have been estimated (Futselaar et al., 1993; Wiesner et al., 1994; Sethi and Wiesner, 1995; Adham et al., 1996; Gere, 1997; Chellam et al., 1998); however, most cost studies have been based on water treatment applications. Recently, a cost estimation model has been applied to agricultural systems (Singh and Cheryan, 1998). Crossflow microfiltration with flux enhanced technology has been shown to be an economical process for the food industry. For instance, Ramirez and Davis (1998) calculated the cost for operating a rapid back-flushing microfiltration system and showed a lower cost compared to conventional methods.

Although two-phase flow microfiltration successfully reduces the fouling problem, a preliminary economic analysis is still required to determine the benefit of two-phase flow microfiltration. Several cost models which were developed to calculate the costs for conventional microfiltration (Pickering and Wiesner, 1993; Wiesner et al., 1994), can be modified for two-phase flow microfiltration. Similar to conventional microfiltration, the economic analysis of two-phase flow microfiltration involves capital and operating costs.

## 2.10.1 Capital costs

Capital costs represent the investment required to install a cross-flow

microfiltration unit. This investment includes membrane costs, construction costs, and installation costs. Capital costs are usually divided into non-membrane costs  $(C_{NM})$  and the initial cost of the membrane modules  $(C_{MM})$ .

# Non-membrane costs

Pickering and Wiesner (1992) developed a mathematical model which assumed that non-membrane costs were correlated with the number of installed membrane modules, and Ramirez and Davis (1998) used this cost model to calculate non-membrane costs for a rapid back-flushing microfiltration. However, the model developed by Pickering and Wiesner was only for water treatment plants. Non-membrane costs for building a cheese whey microfiltration process include all equipment and facilities necessary to support the use of membranes, such as pumps, monitoring equipment, valves, pipes, etc. Non-membrane costs ( $C_{NM}$ ) are the sum of these expenses.

## Membrane cost

The number of required membrane modules (N<sub>MM</sub>) is calculated by:

$$N_{MM} = int \left[ \frac{Q_{req}}{J \bullet A_{MM} \bullet T_F} \right] + 1$$
(13)

where Q<sub>rea</sub> = volumetric treatment rate (plant capacity), m<sup>3</sup>/h

 $A_{MM}$  = surface area of each membrane module, m<sup>2</sup>

J = permeate flux,  $L/(m^2 \cdot h)$ 

int = integer function

 $T_F$  = filtration time, h

The capital costs attributable to the initial purchase of membrane modules are calculated as the product of the number of membrane modules required and the cost of each membrane module.

$$C_{MM} = C_{PM} \bullet N_{MM} \tag{14}$$

where  $C_{MM}$  = initial cost of the membrane modules, \$

C<sub>PM</sub> = cost of one membrane module, \$

The total capital costs,  $C_{TC}$  (\$), can be calculated by:

$$C_{TC} = C_{NM} + C_{MM} \tag{15}$$

# 2.10.2 Operating costs

Operating costs represent the expenses associated with energy consumption, labor, chemicals, and maintenance.

## Energy cost

Energy costs are calculated from energy requirements for cleaning procedures, air compressors, and pumping the feed solution. The sum of these energy requirements, multiplied by the cost of energy, yields the total energy costs for the membrane unit.

$$C_{TE} = \left(\frac{E_{FS} + E_{CP}}{\eta_1} + \frac{E_{AC}}{\eta_2}\right) \bullet C_{KW}$$
(16)

where  $C_{TE}$  = total energy costs, \$

 $E_{FS}$  = required energy for pumping feed solution  $E_{CP}$  = required energy for cleaning procedures  $E_{AC}$  = required energy for air compressor  $C_{KW}$  = electricity cost per kilowatt-hour, \$/(kw-h)  $\eta_1$ ,  $\eta_2$  = pump efficiency

# **Chemical cost**

The cost of adding cleaning reagents to clean the membrane is calculated from the chemical dosage, the amount of cleaning fluid, and the bulk cost of the chemical. Because the chemical cost depends on the cleaning procedures, it can be highly varied.

$$C_{cc} = D_c \cdot V_c \cdot M_w \cdot C_{BC} \cdot N_c \tag{17}$$

where  $C_{cc}$  = chemical cost, \$

 $D_c$  = chemical dosage for cleaning membrane, N

 $V_c$  = amount of cleaning fluid, m<sup>3</sup>

M<sub>w</sub> = molecular weight of chemical, kg/mole

C<sub>BC</sub> = bulk cost of chemical, \$/kg

N<sub>c</sub> = number of cleaning cycles in one year

## Maintenance costs

The maintenance costs include membrane replacement costs and labor costs. Based on the membrane life-time provided by the membrane manufacturer, membrane replacement costs can be modeled as a constant operating cost by assuming that all membranes are replaced at fixed intervals. Depending on the application, the labor cost of operating a two-phase flow microfiltration system can be either a negligible or dominate cost. These annual maintenance costs can be calculated by the following equations:

$$C_{MR} = \frac{C_{MM}}{T_{Life}} + C_{Parts}$$
(18)

$$C_{Labor} = W_{Labor} \cdot N_{Labor} \tag{19}$$

where  $C_{MR}$  = cost of membrane replacement, \$

C<sub>Parts</sub> = cost of parts replacement, \$

T<sub>Life</sub> = membrane life time, yr

C<sub>Labor</sub> = labor cost, \$

W<sub>Labor</sub> = annual labor wage, \$

N<sub>Labor</sub> = number of laborers

The total annual operating costs,  $C_{TO}$  (\$), can be calculated by:

$$C_{TO} = C_{TE} + C_{CC} + C_{MR} + C_{Labor}$$
(20)

# **CHAPTER 3**

# MATERIALS AND METHODS

#### 3.1 Preparation of solutions

Two types of solutions, fungal biomass solutions and cheese whey solutions, were used in this research. Fungal biomass solutions were used for preliminary study of cross-flow microfiltration. The effect of particle concentration on the permeate flux was also studied. Both fungal biomass solutions and cheese whey solutions were used to study the characteristics of conventional cross-flow microfiltration as well as two-phase flow microfiltration. In addition, cheese whey solutions were used to perform the energy studies for single-phase and two-phase cross-flow microfiltration systems. The operating parameters, such as liquid flow rate, transmembrane pressure, and air percentage, were optimized based on the permeate flux and energy requirement data.

# 3.1.1 Fungal biomass solutions

*Pythium irregulare* was maintained on corn meal agar (Difco Laboratories, Detroit, MI) and transferred to new agar slants every three months. Cultivation of *Pythium irregulare* was achieved in a 2% glucose medium supplemented with 0.5% yeast extract (Difco Laboratories) and 0.1 % KPO<sub>4</sub> with an adjusted pH of 6.5. The cultivation was accomplished aerobically using an air-lift bioreactor (Fig. 7). The incubation temperatures were set at 20, 24 and 28°C, and the incubation time was



Figure 7. Schematic diagram of air-lift bioreactor.

2 days. Both the incubation temperature and pH were monitored and controlled. After the cultivation was completed, the biomass solution was transferred to a storage container and stored in a refrigerator (4°C).

Three fungal biomass solutions (FBS1, FBS2, and FBS3) were used for microfiltration studies. Each of them was fermented in an air-lift bioreactor under different conditions; however, all of these biomass solutions contained fungi, yeast extract, and other nutrients. Total solids contents of these biomass solutions were verified by drying in an vacuum oven (Baxter<sup>®</sup> Scientific Products, Model no. N7595-1) at 70°C overnight (AOAC, 1990). After cooling the samples in a desiccator, they were re-weighted. Then total solids contents (TS%) were determined as:

$$TS(\%) = \frac{Mass (after drying)}{Mass (before drying)} \times 100\%$$
(21)

Table 1 shows the operating parameters for fungal biomass cross-flow microfiltration studies.

#### 3.1.2 Cheese whey solutions

Cheese whey solutions were provided by Purity<sup>®</sup> Dairy (Nashville TN). The pH values of these cheese whey solutions was approximately 4.3, and the total solids content was 6.3%. The protein, fat, and ash contents were determined as 0.75%, 0.02%, and 0.6% respectively (Martinez-Hermosilla, 1999). These cheese

Table 1.	Operating parameters for fungal biomass cross-flow microfiltration
	studies.

Parameter	Unit	Value		
		FBS1	FBS2	FBS3
Total solids content	%	0.34	1.15	0.5
Temperature	°C	20	20	20
Initial feed volume	Ĺ	8.2	10	10
Transmembrane pressure (TMP)	kPa	62	62	62
Liquid flow rate	L/min	8	9.7	9.7
Air flow rate	L/min	7.2	7.2	7.2

whey solutions were well mixed then transferred into 10 L buckets and stored in a freezer (-18°C). Each bucket of cheese whey solution was thawed at room temperature two days before running the filtration experiments.

Microfiltration was used as a pretreatment to remove lipids before ultrafiltration. The permeate of microfiltration was collected and frozen (-18°C) for further ultrafiltration research involving the production of whey protein concentrates.

Table 2 shows the operating parameters for cheese whey cross-flow microfiltration studies.

## 3.2 Experimental setup

The experimental apparatus used in this research is shown in Fig. 8. The elements in the experimental apparatus were connected with Tygon<sup>®</sup> B-44-4X tubing (inside diameter 0.953 cm [ $\frac{3}{5}$  in.]). The cross-flow microfilter was manufactured by A/G Technology Corporation<sup>®</sup> (Needham, MA) and was made from polysulfone. The microfilter tube bundle (36 tubes) had an outside diameter of 60 mm and a length of 650 mm. The inside diameter of each tube was 3 mm. The pore size and the total cartridge membrane area were 0.1 µm and 0.15 m<sup>2</sup> respectively. The schematic diagram of the cross-flow microfilter is shown in Fig. 9.

Two pressure gauges (Metek<sup>®</sup> model no. 1X682) were used to measure the inlet and outlet pressure (P<sub>inlet</sub> and P<sub>outlet</sub>) of the microfilter. The transmembrane pressure (TMP) was defined in equation 10. Because the permeate flow was

Parameter	Unit	Value		
рН		4.2		
Total solids content	%	6.3		
Temperature	°C	20		
Initial feed volume	L	10		
Transmembrane pressure (TMP)	kPa	2.1 ~ 112.3ª		
Liquid flow rate	L/min	3 ~ 20 <sup>b</sup>		
Air flow rate	L/min	0 ~ 5.6° (0 ~ 40% at TMP = 62 kPa)		

Table 2. Operating parameters for cheese whey cross-flow microfiltration studies.

<sup>a</sup> This range was used for a test to determine the effect of transmembrane pressure. All other tests used a transmembrane pressure of 62 kPa.

<sup>b</sup> This range was used for a test to determine the effect of liquid flow rate. All other tests used a liquid flow rate of 10 L/min.

<sup>c</sup> This range was used for a test to determine the effect of air flow rate.



1. inlet pressure gauge ( $P_{inlet}$ ); 2. outlet pressure gauge ( $P_{outlet}$ ); 3. cross-flow microfilter; 4. back pressure control valve; 5. cheese whey reservoir (20 L); 6. foam collector; 7. permeate collector; 8. peristaltic pump; 9. air pump; 10. air flow rate control valve; 11. check valve; 12. pressure gauge ( $P_a$ ); 13. rotometer flow control valve; 14. rotometer

Figure 8. Schematic diagram of experimental set-up.



**Cross-flow Microfilter** 

Figure 9. Schematic diagram of cross-flow microfilter.

exposed to the atmosphere, the permeate was assumed to have zero gauge pressure. The transmembrane pressure was therefore calculated as the average of the sum of the inlet and outlet pressures. A back-pressure control valve downstream from the membrane was used to obtain the desired transmembrane pressure. Normally, the transmembrane pressure was maintained at 62 kPa.

A variable-speed peristaltic pump (MasterFlex<sup>®</sup> model no. 7585-30) with Norprene<sup>®</sup> food tubing (Norton<sup>®</sup>, model no. 06402-90) was used to pump the solutions through the microfilter. This peristaltic pump was calibrated first to measure the liquid flow rate at each pump speed. The liquid flow rate was calculated by measuring the volumetric flow of liquid over time. Then the desired liquid flow rate could be achieved by setting the pump speed to the corresponding reading. The maximum capacity of this peristaltic pump was 30 L/min. In the microfiltration studies, the pump was normally operated at 10 L/min. Due to the high pump speed, the Norprene<sup>®</sup> tubing was subjected to excessive wear and needed to be replaced after several runs. With different discharge pressures, the pump calibration procedure needed to be repeated.

An air pump (Gast<sup>®</sup> model no. DOA-P104-AA) was used to pump air into the stream to achieve a two-phase flow. The air tubing was connected to the liquid tubing by using a 'Y' connector. A check valve was used to prevent the liquid from backing up into the air pump. The air flow rate was controlled by a control valve and monitored by a rotometer (Cole-Parmer<sup>®</sup> model no. N034-39). Air pressure was also measured by an air pressure gauge.
Air pressure ( $P_{Air}$ , kPa), air flow rate ( $V_{air}$ , L/min), and two-phase flow pressure ( $P_{TPF}$ , kPa) were required information to calculate the air percentage in the flow. If the liquid flow rate was measured as  $V_{Liquid}$  (L/min), the air flow rate in the liquid stream can be calculated by applying the ideal gas assumption:

$$P_{Air} \bullet V_{Air} = P_{TPF} \bullet V_{TPF}$$
(22)

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where  $V_{TPF}$  = the air flow rate in the two-phase flow, L/min. Then the air percentage in the two-phase flow can be calculated as:

$$Air\% = \frac{V_{TPF}}{V_{TPF} + V_{Liquid}} \times 100\%$$
(23)

A 110 V kilowatt-hour meter was used to measure the energy consumption for both the peristaltic pump and air pump. An infrared sensor and a digital counter were connected to the kilowatt-hour meter to collect precise readings. This kilowatt-hour meter was calibrated by the service department of Tennessee Valley Authority.

After each experiment, the membrane needed to be cleaned. The cleaning procedures included back-washing with permeate, water flushing, and chemical treatment. When the filtration process was completed, permeate solution was pumped back to back-flush the membrane and the back-flushing pressure was not allowed to exceed 34 kPa. After 2 min of back-flushing, warm water (50°C) was recycled through the membrane for 20 min. Then some chemical reagents were

added to clean the membrane depending on the application. For fungal biomass microfiltration, 50 ppm chlorine (50°C) was used to sanitize the membrane. For cheese whey microfiltration, 0.5 N,  $\text{NaOH}_{(aq)}$  (50°C) was used to clean oils and fats. These chemical reagents were recycled through the membrane for 40 min. The membrane was then washed with warm water for another 40 min. These cleaning procedures were recommended by the membrane manufacturer (A/G Technology Corporation<sup>®</sup>).

#### 3.3 Velocity head study

Clean water was pumped through the microfilter at rates from 3 L/min to 17 L/min. The back-pressure control valve was adjusted to keep the apparent transmembrane pressure at 62 kPa. The experiment was repeated with the transmembrane pressure corrected for the velocity head effect. The cross-sectional area in the large tubes where the pressure gauges were placed was 0.71 cm<sup>2</sup> (d=0.953 cm) and the total cross-sectional area in the 36 membrane tubes was 2.55 cm<sup>2</sup> (d=0.3 cm) (See Fig. 9). Therefore, the flow velocity was reduced when the liquid was pumped into the membrane tubes. For example, at 16 L/min, the flow velocity in the feed and discharge tubing was 3.74 m/s, which was reduced to 1.05 m/s in the membrane tubes. Based on equation 12, the velocity head effect increased the pressure in the membrane tube by 13 kPa. The apparent transmembrane pressure was thus corrected with a 13 kPa reduction.

The entire procedure was repeated with the cheese whey solution. The

cheese whey solution was pumped through the membrane without air. According to equation 12, the apparent transmembrane pressure was reduced corresponding to the liquid velocity. The corrected transmembrane pressure was set at 62 kPa. The permeate was recycled into the reservoir to maintain a constant feed concentration. Steady-state was obtained rapidly, so all data were collected after 20 min filtration time. All the experiments were performed at room temperature.

#### 3.4 Effect of two-phase flow on cross-flow microfiltration

Both fungal biomass solutions and cheese whey solutions were used to study the effect of the two-phase flow on microfiltration. The air flow rate control valve (Fig. 8) and the rotometer flow control valve were used to control the amount of injected air. Based on the air pressure, air flow rate, liquid flow rate, and transmembrane pressure, the percentage of injected air could be calculated (equation 23). The permeate was measured, and then recycled into the reservoir with retentate to maintain a constant feed concentration. Because mixing air into the liquid flow generated foam, a closed container was connected to the reservoir to be a foam collector (Fig. 8). The temperature of the reservoir was maintained at room temperature.

For fungal biomass solutions, the liquid flow rates for FBS1, FBS2, and FBS3 were 8.0 L/min, 9.7 L/min, and 9.7 L/min, respectively. The air flow rate was 7.2 L/min (32~37%). The corrected transmembrane pressure ranged from 55.2 to 58.6 kPa. The two-phase flow was generated by intermittent introduction of air and

continual mixing of air. For intermittent introduction of air, the air pump was turned on and off periodically. At the beginning, the fungal biomass solutions were filtered without air for 25 min before the air pump was turned on. After mixing air bubbles for 5 min, the air pump was turned off. The fungal biomass solutions were filtered without air for another 25 min, followed by another 5 min period of air injection. These procedures were repeated four times. For continual mixing, the air bubbles were continuously injected into the fungal biomass solutions. In one experiment, the air was continually injected after filtration for 90 min without air.

For cheese whey solutions, the liquid flow rate and the corrected transmembrane pressure were set at 10 L/min and 62 kPa, respectively. Air was pumped into the stream at flow rates varying from 0 L/min to 5.6 L/min, which corresponded to 0% to 40% air. Because the process reached a steady-state rapidly, the permeate flow data were taken after 10 min operating time. The permeate was also recycled into the reservoir to maintain a constant feed concentration.

#### 3.5 Economic analysis

The total costs for a cheese producer to build a cross-flow microfiltration system to pretreat cheese whey were estimated. It was assumed that the cheese plant produces about 50,000 gallons of cottage cheese whey per week (189 m<sup>3</sup>/week). Instead of using back-flushing, two-phase flow microfiltration was introduced to reduce the fouling and improve the performance of the membrane.

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The membrane information, such as price, life-time, and total area of each membrane module, was provided by A/G Technology Corporation<sup>®</sup>.

Both capital and operating costs were calculated based on the capacity of the plant. The annual capital costs, including non-membrane and membrane costs, were calculated by equation 15. The operating costs, including energy, chemical, and maintenance costs, were calculated individually. The total energy consumption for both the single-phase and two-phase flow microfiltration was estimated based on laboratory scale energy measurements. In single-phase flow microfiltration, the energy consumption was measured with a series of liquid flow rates. In two-phase flow microfiltration, the energy consumption was measured for both the liquid pump and the air pump. Based on the steady-state permeate flux data, the processing time and energy requirement for collecting 1000 L permeate liquid were calculated. The frequency of cleaning membrane modules was assumed to be weekly, and the procedures were identical to the cleaning steps used in the lab-scale microfiltration. Based on the capacity of membrane modules and cleaning frequency, the amount of required chemical (NaOH) was calculated. In addition, the costs of energy consumption for cleaning membranes plus labor and membrane replacement costs were estimated as well. Then the total annual operating costs were calculated by combining these costs together.

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# **CHAPTER 4**

# **RESULTS AND DISCUSSION**

#### 4.1 General characteristics of fungal biomass microfiltration

A typical permeate flux decline for the three biomass solutions (FBS1, FBS2, and FBS3) is shown in Fig. 10. The transmembrane pressure was adjusted to 62 kPa and kept constant by adjusting the back pressure control valve. The liquid flow rate was set at 10 L/min. Although the permeate flux rates were different, they all reached the saturated (pseudo steady-state) stage. The permeate flux declined drastically at the initial stage due to membrane fouling then reached a pseudo steady-state stage. The term "steady-state" meant that the permeate flux reached a stable level even though some measurement variation still existed. The steady-state flux rate at which particles were added to the cake, became equal to the rate at which they were removed, and the cake layer thickness ( $\delta$ ) was constant. Because the retentate was recycled into the reservoir, the bulk concentration of the fungal biomass solution increased. Therefore the permeate flux declined at a much slower rate at the steady-state stage.

Although the particle concentration has influence on the permeate flux, the effect was not clear in fungal biomass microfiltration. The physical properties (like viscosity and particle shape) of these fungal biomass particles were quite different from each other; thus, the particle concentration effect was hard to analyze.



Figure 10. The permeate flux decline of FBS1, FBS2, and FBS3 without air bubbles.

# 4.2 General characteristics of cheese whey microfiltration

A typical permeate flux decline curve for cheese whey microfiltration is shown in Fig. 11. The transmembrane pressure was adjusted to 62 kPa and the liquid flow rates were set at 5 L/min, 10 L/min, and 20 L/min respectively. The reduction in flux was attributed to membrane fouling. Because cheese whey contains small amounts of fat and lipids which easily block the membrane pores, the permeate flux declined drastically at the initial stages. This situation was very similar to fungal biomass microfiltration. According to the flux decline curve, the cheese whey solution fouled the micromembrane in the first 10 min in all three cases, and then a steady-state flux was reached. To track the fouling, the permeate flux data were taken every five minutes and averaged. Thus, the variations in the first 10 min of operation were more obvious. For the 20 L/min liquid flow rate, the permeate flux took about 10 min to reach steady-state, but for the 5 L/min flow, the permeate flux reached a steady-state almost immediately.

The steady-state permeate flux data for 5, 10, and 20 L/min liquid flow rates were 7.5, 11, and 16 L/(m<sup>2</sup>·h) respectively. According to these data, increasing the liquid flow rate resulted in a higher steady-state permeate flux. However, this was true only when the filtration system was in the mass transfer controlled region. If the liquid flow rate was increased above a certain level, the filtration system eventually would become pressure controlled. In the pressure controlled region, the effect of increasing liquid flow rate on permeate flux becomes minimal.



Figure 11. Typical permeate flux decline for cheese whey cross-flow microfiltration (Corrected transmembrane pressure = 62 kPa).

#### 4.3 Operating parameter study

It is known that a higher transmembrane pressure results in a higher permeate flux. Clean water was used to demonstrate this phenomenon, and the result is shown in Fig. 12. Because clean water did not foul the membrane, the membrane resistance was constant and the relationship between the transmembrane pressure and the permeate flux was linear. According to equation 12, the pressure change is proportional to the velocity squared; therefore, it was necessary to correct the pressure effect caused by the velocity change to see the true effect of the liquid velocity on the permeate flux. The liquid velocity was determined by dividing the volumetric flow rate by the cross-sectional area. In this experiment, the total cross-sectional area in the membrane tubes was 3.6 times larger than the cross-sectional area where the pressure gauges were placed (Fig. 9). Therefore, the fluid in the membrane tubes was at a lower velocity than at the pressure gauges, so the transmembrane pressure was higher than that indicated by the pressure gauges. The pressure change curve caused by the liquid flow rate is shown in Fig. 13. When the liquid flow rate was low, the pressure change caused by the velocity change was negligible. When the liquid flow rate was increased, the pressure change became more significant. In most cases, the operating transmembrane pressure was set at 62 kPa. When the liquid was pumped at 10 L/min, the pressure change caused by the velocity effect was only 4.8 kPa. However, when the liquid was pumped at 16 L/min, the pressure change was 13 kPa. This pressure change caused by the velocity head effect was taken into



Figure 12. The transmembrane pressure effect on the permeate flux of clean water (Liquid flow rate = 10 L/min).



Figure 13. Pressure change caused by the liquid velocity head effect.

account and apparent transmembrane pressure was adjusted accordingly.

Fig. 14 shows the effect of velocity head on the permeate flux for the clean water system. The operating transmembrane pressure was set at 62 kPa. Without considering the velocity head effect, the permeate flux increased with an increased liquid flow rate. However, there was no fouling problem with clean water, thus, the permeate flux should not have changed under a constant transmembrane pressure. After correcting for the velocity head effect, the permeate flux did remain constant (230 L/m<sup>2</sup>·h). However, under the same operating conditions (62 kPa, 10 L/min), the maximum permeate flux for cheese whey microfiltration was only 32 L/m<sup>2</sup>·h (see Fig. 11). The difference was caused by the fouling. Because cheese whey solutions contain fat and lipids, they fouled the micromembrane very quickly and the permeate flux dropped dramatically.

The velocity head effect affected not only the clean water system but also the cheese whey filtration. Fig. 15 shows the effect of velocity head on the cheese whey microfiltration. The operating apparent transmembrane pressure was also set at 62 kPa. Similar to the clean water system, without correcting for the velocity head effect, a higher permeate flux was achieved. The permeate flux increased about 29% when the liquid flow rate was doubled. After correcting for the velocity head effect, the permeate flux only increased 20% when the liquid flow rate was doubled. However, when the liquid flow rate was slower than 10 L/min, the pressure difference caused by the velocity head effect was very small, so it was difficult to correct for this small amount of pressure change. Thus, the two



Figure 14. Velocity head effect on the permeate flux of clean water (Apparent TMP = 62 kPa).



Figure 15. Velocity head effect on cheese whey microfiltration (Apparent TMP = 62 kPa).

permeate flux curves merged together at low liquid flow rate.

Fig. 15 also revealed that increasing liquid flow rate improved the permeate flux. The steady-state permeate flux at 10 L/min liquid flow rate and 62 kPa was only 8 L/(m<sup>2</sup>·h). According to Fig. 15, increasing liquid flow improved the permeate flux to 8.8 L/(m<sup>2</sup>·h) and 9.6 L/(m<sup>2</sup>·h) with 15 L/min and 20 L/min liquid flow rates, respectively. However, the increased energy requirements to achieve a higher liquid flow which is the biggest drawback. Thus, two-phase flow microfiltration was proposed as an additional flux enhancement method.

The transmembrane pressure effect on cheese whey microfiltration is shown in Fig. 16. The flow rate of cheese whey solutions was set at 10 L/min and kept constant. The permeate flux increased rapidly at lower transmembrane pressures but leveled off at higher transmembrane pressures. The relationship can be explained by the formation of the cake-layer. The cake-layer, built by the cheese whey lipids, created a major resistance to the permeate flux. At low pressure, the permeate flux increased dramatically with increasing pressure. When the pressure became higher, the cake was packed more tightly, which produced more resistance to the permeate flux. As a result, the permeate flux enhancement with increasing transmembrane pressure became less significant.

## 4.4 Effect of two-phase flow on cross-flow microfiltration

The permeate flux can be improved by increasing either the liquid flow rate or the transmembrane pressure. However, both methods require more energy, and



Figure 16. Transmembrane pressure effect on cheese whey microfiltration (Liquid flow rate = 10 L/min).

sometimes the permeate flux improvement is limited. The two-phase flow method has been introduced to improve the permeate flux. Fig. 17 shows the effect of air bubbles intermittently mixed into the FBS1 biomass solution. From 32 to 37% air was used in all experiments to make sure the fouling resistance was significantly reduced. When the air bubbles were mixed into the liquid, the permeate flux increased. Once the air was turned off, the permeate flux declined very quickly. Air bubbles cleaned the membrane surface, allowing more permeate to go through. After the air was turned off, cake formation occurred and the permeate flux declined again. Therefore, intermittent introduction of air is not nearly as effective as continual mixing of air, as is discussed below.

In the next experiment, air bubbles were mixed into the FBS1 biomass solution after 90 min of filtration without air (Fig. 18). Once the air bubbles hit the membrane, the permeate flux increased dramatically, as in the previous case. Air was mixed continuously from that point on. The initial shock of the air introduction reduced the cake thickness significantly. The flux then declined again as the cake-layer was reformed at a reduced steady state thickness. The flux at this point was approximately 100% greater than the flux before air was added. However, after 90 min of filtration without air, the cake-layer was formed completely. It became more difficult to wash this cake out.

When air was continuously mixed into the biomass solution under the same operating conditions, a higher permeate flux was achieved. The effect of air bubbles is shown in Fig. 19 to 21 for the three types of biomass solutions. Air



Figure 17. The effect of intermittent mixing of air bubbles into FBS1 biomass solution (37% Air, Initial  $C_B = 0.34$  g/L, Flow Rate = 8.0 L/min).







Figure 19. The permeate flux decline of FBS1 biomass solution (37% Air, Initial  $C_B = 0.34$  g/L, Flow Rate = 8.0 L/min).



Figure 20. The permeate flux decline of FBS2 biomass solution (32% Air, Initial  $C_B$  = 1.15 g/L, Flow Rate = 9.7 L/min).



Figure 21. The permeate flux decline of FBS3 biomass solution (32% Air, Initial  $C_B$  = 0.5 g/L, Flow Rate = 9.7 L/min).

bubbles maintained an increase in the permeate flux of 120%, 45%, and 40% for FBS1, FBS2, and FBS3 respectively, after one hour operation time. The difference in the increase in efficiency is due to the varying physical properties of these biomass solutions. Because the FBS1 solution resulted in the greatest fouling with no air present, it had the greatest potential for flux increase. The difference of permeate flux increase between Fig. 18 and Fig. 19 was due to the starting point of air injection.

Air bubbles provided a driving force to shear the cake-layer from the membrane. When air bubbles were passed through the microfilter to wash out those particles, the permeate flux was maintained at a significantly higher level. In other words, air bubbles can reduce the cake thickness to increase the permeate flux. However, the permeate flux still reached a saturated stage though air bubbles were continuously mixed into the biomass solution. Once the permeate flux drops to some predetermined levels, it may be better to clean the membrane instead of running the two-phase filtration process at a relatively low permeate flux rate.

The two-phase flow method has been shown to be an effective method to prevent and reduce the fouling. The question was how much injected air was required to improve the permeate flux. Fig. 22 shows the permeate flux improvement with two-phase flow for cheese whey microfiltration. The air percentage in the mixture was varied from 0% to 40% (v/v). When 5% air was mixed, the permeate flux increased about 50%. With 30% air, the permeate flux increased about 78%. It was interesting to see that a small amount of air improved





the permeate flux significantly. This result was similar to that obtained by Cui (1993), Cui and Wright (1994), Cui and Wright (1996), and Bellara et al. (1996). A small amount of air was enough to provide the necessary shear force to shear the cake out off the membrane. Injecting more air could result in a higher permeate flux, but this requires additional energy. The permeate flux improvement needs to be optimized with respect to energy requirements.

## 4.5 Economic analysis

#### 4.5.1 Energy study

Tables 3 and 4 show the energy requirements for cheese whey cross-flow microfiltration under a constant transmembrane pressure operation. These results indicated how much time (h) and how much energy (kJ) were required to collect 1000 L permeate under steady-state conditions. Table 3 shows the effect of injecting air into cheese whey solutions pumped at 10 L/min. Injecting 5% air into the stream shortened the operating time by 41% and reduced the total energy requirements by 26%. On the other hand, injecting 40% air reduced the processing time 52% time but increased the energy requirements by 44%. As mentioned previously, increasing the liquid flow rate could also improve the microfiltration performance. Table 4 shows the comparison of energy requirements for several different liquid flow rates. When the liquid flow rate was increased from 10 L/min to 15 L/min, the processing time was reduced 9% but required 28% more energy. Likewise, when the liquid flow rate was doubled to 20 L/min, the processing time

# Table 3. Energy and processing time requirements for two-phase flow cheese whey microfiltration (based on air percentage)

Liquid flow rate = 10 L/min

Parameter	Unit			Value		
Air percentage	%	0	5	10	20	40
Permeate flux	L/m² h	8.0	13.5	14.2	14.6	16.7
Permeate flow rate	L/h	1.20	2.03	2.13	2.19	2.51
Energy required for one hour operation	J	110	137.5	165	220	330
Time required for collecting 1000 L permeate	h	833	494	470	457	399
Energy required for collecting 1000 L permeate	kJ	92	68	78	101	132
Time saved	%	Baseline	41	44	45	52
Energy required	%	Baseline	-26*	-15*	+10	+44

\* require less energy

Parameter	Unit		Value	
Liquid flow rate	L/min	10	15	20
Permeate flux	L/m² h	8.0	8.8	9.6
Permeate flow rate	L/h	1.20	1.32	1.44
Energy required for one hour operation	J	110	155	225
Time required for collecting 1000 L permeate	h	833	758	694
Energy required for collecting 1000 L permeate	kJ	92	117	156
Time saved	%	Baseline	9	17
Energy required	%	Baseline	+28	+70

 Table 4. Energy and processing time requirements for cheese whey microfiltration (based on liquid flow rate).

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was reduced 17% but required 70% more energy. Based on both processing time and energy requirements, two-phase flow was a more efficient means of improving membrane performance when compared to simply increasing the liquid flow. A low amount of air (5~10%) was practical for most applications. Injecting more air into the stream saved additional time, but the total energy requirement was increased. Also, foaming problems were more predominant as the amount of air was increased. The comparison of energy and processing time requirements for cheese whey microfiltration is shown in Figs. 23 and 24.

## 4.5.2 Cost estimation

Fig. 25 shows the schematic diagram of a scaled-up cheese whey microfiltration system. This design includes the basic elements for two-phase flow microfiltration. The capacity of the cheese manufacturing plant is assumed to be 50,000 gallons of cottage cheese (189 m<sup>3</sup>) per week. The number of required membrane modules was determined by the steady-state permeate flux and plant capacity (Equation 13). Four types of micromembranes with 0.1 µm pore size were considered in cost estimation, and the total membrane area, internal diameter of tubules, and price information are listed in Table 5. The operating parameters were assumed to be the same as in the lab-scale microfiltration. The transmembrane pressure and the liquid flow rate for each membrane module remained at 62 kPa and 10 L/min, respectively. The air percentage for two-phase flow microfiltration also remained 5%. The non-membrane costs, including pumps ( $C_{pump}$ ), valves and



Figure 23. Energy and processing time requirements (based on air percentage) for two-phase flow cheese whey microfiltration (for collecting 1000 L cheese permeate solution; Liquid flow rate = 10 l/min).



Figure 24. Energy and processing time requirements (based on liquid flow rate) for two-phase flow cheese whey microfiltration (for collecting 1000 L cheese permeate solution).



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Figure 25. Schematic diagram of a scaled-up cheese whey microfiltration.

Internal diameter of tubule (mm)	Pore size (µm)	Membrane area (m²)	Price
3	0.1	4.2	\$3,500
2	0.1	5.6	\$3,600
1	0.1	8.8	\$4,000
0.5	0.1	13	\$5,000

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Table 5.	Physical properties of membrane modules (provided by A/G
	Technology Corporation <sup>®</sup> ).

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piping ( $C_{VP}$ ), installation and parts ( $C_{IP}$ ), and monitoring equipment ( $C_{ME}$ ), were estimated. The membrane cost was calculated based on the number of required membrane modules which is shown in Table 6.

Tables 7 and 8 show the cost estimates, including capital costs and annual operating costs, for building and operating single-phase and two-phase flow microfiltration systems. The major costs were membrane cost and labor costs. Appendix A and B show examples for calculating capital costs and annual operating costs for single-phase and two-phase flow microfiltration systems, respectively. Increasing the membrane area significantly reduced the membrane cost, but the remainder of the costs basically remained the same. The capital costs for building single-phase and two-phase flow microfiltration with four types of a micromembranes are shown in Fig. 26. Capital costs decreased with increasing membrane area because the number of required membrane modules was reduced. In addition, the two-phase method also significantly reduced the capital costs. Although the membrane with an area of 13 m<sup>2</sup> provided the lowest cost, the 0.5 mm internal diameter of tubules was too narrow for cheese whey applications. Therefore, the 8.8 m<sup>2</sup> membrane was the ideal membrane to choose. Fig. 27 shows that the membrane cost is the major capital cost for building a scaled-up microfiltration system.

Fig. 28 shows the annual operating costs for running single-phase and twophase flow microfiltration systems with four types of micromembranes. Increasing the membrane area reduced operating costs due to lower chemical and membrane

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Membrane area (m²)	Single-phase flow microfiltration	Two-phase flow microfiltration	
4.2	40	24	
5.6	30	18	
8.8	19	12	
13	13	8	

 Table 6. Number of required membrane modules for single-phase and two phase flow microfiltration.

Table 7.	Cost estimation for single-phase flow microfiltration		
	(The capacity of cheese manufacturing plant = 189 m <sup>3</sup> /week)		

Membrane area	4.2 m <sup>2</sup>	8.8 m <sup>2</sup>
Capital costs	Dollars	Dollars
Non-membrane costs	\$15,000	\$15,000
Membrane cost	\$140,000	\$76,000
Total	\$155,000	\$91,000
Membrane area	4.2 m <sup>2</sup>	8.8 m <sup>2</sup>
Annual operating costs	Dollars	Dollars
Energy costs	\$10,500	\$10,500
Chemical cost	\$5,200	\$2,600
Membrane replacement costs	\$29,000	\$16,200
Labor costs	\$70,000	\$70,000
Total	\$114,700	\$99,300
Table 8.	Cost estimation for two-phase flow microfiltration	
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	(The capacity of cheese manufacturing plant = 189 m <sup>3</sup> /week).	

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Membrane area	4.2 m <sup>2</sup>	8.8 m <sup>2</sup>
Capital costs	Dollars	Dollars
Non-membrane costs	\$17,000	\$17,000
Membrane cost	\$84,000	\$48,000
Total	\$101,000	\$65,000
Membrane area	4.2 m <sup>2</sup>	8.8 m <sup>2</sup>
Annual operating costs	Dollars	Dollars
Energy costs	\$11,460	\$11,460
Chemical cost	\$3,640	\$2,080
Membrane replacement costs	\$17,800	\$10,600
Labor costs	\$70,000	\$70,000
Total	\$102,900	\$94,140



Total area of each membrane module (m<sup>2</sup>)

Figure 26. Capital costs for a scaled-up microfiltration (The capacity of cheese manufacturing plant = 189 m<sup>3</sup>/week).



Figure 27. Capital costs analysis for a scaled-up microfiltration (with 8.8 m<sup>2</sup> micromembranes).



Total Area of Each Membrane Module (m<sup>2</sup>)



replacement costs. However, labor costs remained the same, and these were the major costs for operating a microfiltration process. Although the two-phase flow method reduced the annual operating costs for all membrane sizes, the amount of saving was less significant comparing to the savings in capital costs. Fig. 29 shows the labor costs are the major costs for operating a microfiltration process.

Fig. 30 shows the cost saving percentage for building and operating a twophase microfiltration. According to the results of the cost estimates, the two-phase flow microfiltration with an 8.8 m<sup>2</sup> membrane area was the most economical process for pretreating cheese whey solutions. The capital costs saving for using 8.8 m<sup>2</sup> micromembranes was 28.6% and the annual operating costs saving was 5.2%. The economic analysis can be improved by collecting more information about the permeate flux under higher liquid flow rates.



Single-phase microfiltration:

Two-phase microfiltration:

5



Figure 29. Annual operating costs analysis for a scaled-up microfiltration (with 8.8 m<sup>2</sup> micromembranes).



Total Area of Each Membrane Module (m<sup>2</sup>)

Figure 30. Cost saving percentage for building and operating a two-phase flow microfiltration compared to single-phase flow microfiltration.

### **CHAPTER 5**

#### CONCLUSIONS

The transmembrane pressure and liquid flow rate were two key factors determining the performance of cross-flow microfiltration. The permeate flux increased with increasing transmembrane pressure. To keep the transmembrane pressure constant when velocity increased, the pressure change caused by the velocity head effect needed to be considered, and the apparent transmembrane pressure was reduced to keep the actual transmembrane pressure constant. The velocity head effect became more significant with increasing liquid flow rate. On the other hand, the permeate flux also increased with increasing liquid flow rate. Without using the two-phase flow method, the permeate flux increased 20% when the liquid flow rate was doubled. Increasing the liquid flow rate eventually moved the system from the mass transfer controlled region to the pressure controlled region. In the pressure controlled region, the effect of increasing liquid flow rate became minimal, and the pressure effect became dominant.

The physical properties of the biomass particles are very important for the microfiltration process. This is especially true for two-phase flow. Particle size, viscosity, and tackiness affect the tendency of the solids to cake and foul the membrane. Because cheese whey contained some fats and lipids, cheese whey solutions fouled the micromembrane very quickly ( $\leq$  15 min). Increasing the transmembrane pressure improved the permeate flux, but the pressure effect on

cheese whey microfiltration was more significant when the transmembrane pressure was less than 30 kPa. Likewise, increasing the liquid flow rate reduced the fouling and resulted in a higher permeate flux.

A series of experiments were performed to determine the effect of two-phase flow on the microfiltration process. Two-phase flow, which was generated by introducing air into the liquid flow, successfully improved the permeate flux of crossflow microfiltration. Air bubbles provided the necessary shear force and pulsation to wash particles off the membrane. The two-phase microfiltration method increased the permeate flux 120%, 45%, and 40% for three different fermented fungal biomass solutions at one hour operating time. For cheese whey microfiltration, the two-phase flow method successfully increased the permeate flux by approximately 50% with only 5% air under 62 kPa transmembrane pressure and 10 L/min liquid flow rate. Continual injection of air was much more effective than was intermittent injection. Although the permeate flux increased with increasing liquid flow, the two-phase flow method was much more efficient in terms of energy and processing time requirements.

The purpose of testing liquid flow rate and air percentage was to establish cost estimates to scale-up the two-phase flow microfiltration process for a cheese manufacturing plant. The costs for two-phase flow microfiltration were significantly less than those for single-phase flow microfiltration. Because two-phase flow improved the permeate flux, fewer membrane modules were required, and the membrane costs were reduced. Increasing the membrane area could also reduce the costs. Because larger membranes are more expensive, the selection of micromembranes needs to be balanced with membrane area and the cost of each membrane. Although two-phase flow reduced total annual costs, the membrane cost and labor cost were still the major expenses for two-phase flow microfiltration.

Future work will involve:

- Setting up a pilot-scale microfiltration unit to examine the permeate flux enhancement from two-phase flow.
- Improving the cost analysis by collecting more information about the effect of operating parameters on the permeate flux.
- Applying automatic control systems to accurately monitor and control the two-phase flow microfiltration process.
- Expanding two-phase flow microfiltration to other liquid food systems.

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**APPENDIXES** 

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## APPENDIX A

Cost estimates for single-phase flow

cheese whey microfiltration

Parameter	Unit	Value
Q	L	189,000
J	L/(m²·h)	8
Amm	m²	8.8
T <sub>F</sub>	h	144
Dc	Ν	0.5
V <sub>c</sub>	L	250
Mw	kg/mol	0.04
Nc	-	52
P <sub>Pump</sub>	hp	10
η	-	0.75
C <sub>BC</sub>	\$/kg	10
C <sub>KW</sub>	\$/(kw-h)	0.103

Table 9. Cost estimates for a scaled-up single-phase flow cheese whey

microfiltration (with 8.8 m<sup>2</sup> membrane).

Total non-membrane costs (C<sub>NM</sub>):

$$C_{NM} = C_{Pump} + C_{VP} + C_{IP} + C_{ME}$$
  
= 3,500+3,000+4,500+4,000=15,000

The required number of membrane modules  $(N_{MM})$  is:

$$N_{MM} = int \left[ \frac{Q_{req}}{J \bullet A_{MM} \bullet T_F} \right] + I$$
$$= int \left[ \frac{189,000}{8 \bullet 8.8 \bullet 144} \right] + I = 19$$

Initial cost of the membrane modules ( $C_{MM}$ ):

$$C_{MM} = C_{PM} \bullet N_{MM}$$
  
= 4,000 • 19 = 76,000

Total capital costs (C<sub>TC</sub>):

$$C_{TC} = C_{NM} + C_{MM}$$
  
= 15,000+76,000=91,000

Energy costs (C<sub>TE</sub>):

$$C_{TE} = \frac{E_{FS} + E_{CP}}{\eta} \cdot C_{KW}$$
$$= \frac{10 \cdot 144 \cdot 52 + 10 \cdot 3 \cdot 52}{0.75} \cdot 0.103 = 10,500$$

Chemical cost (C<sub>cc</sub>):

$$C_{cc} = D_c \cdot V_c \cdot M_W \cdot C_{Bc} \cdot N_c$$
  
= 0.5 \cdot 250 \cdot 0.04 \cdot 10 \cdot 52 = 2,600

Membrane replacement costs ( $C_{MR}$ ):

$$C_{MR} = \frac{C_{MM}}{T_{Life}} + C_{Parts} = \frac{76,000}{5} + 1,000 = 16,200$$

Labor costs (C<sub>Labor</sub>):

$$C_{Labor} = 70,000$$

Total annual operating costs ( $C_{TO}$ ) for single-phase microfiltration:

$$C_{TO} = C_{TE} + C_{CC} + C_{MR} + C_{Labor}$$
  
= 10,500+2,600+16,200+70,000=99,300

### APPENDIX B

Cost estimates for two-phase flow

cheese whey microfiltration

Parameter	Unit	Value
Q	L	189,000
J	L/(m²·h)	13.5
A <sub>MM</sub>	m²	8.8
T <sub>F</sub>	h	144
D <sub>c</sub>	Ν	0.5
V <sub>c</sub>	L	200
M <sub>w</sub>	kg/mol	0.04
Nc	-	52
P <sub>Pump</sub>	hp	7
P <sub>Air</sub>	hp	4
η	· .	0.75
C <sub>BC</sub>	\$/kg	10
C <sub>KW</sub>	\$/(kw-h)	0.103

Table 10. Cost estimates for a scaled-up two-phase flow cheese whey

microfiltration (with 8.8 m<sup>2</sup> micromembranes).

Total non-membrane costs ( $C_{NM}$ ):

$$C_{NM} = C_{Pump} + C_{VP} + C_{IP} + C_{ME}$$
  
= 4,500+3,000+4,500+5,000=17,000

The required number of membrane modules (N $_{\rm MM})$  is:

$$N_{MM} = int \left[ \frac{Q_{req}}{J \bullet A_{MM} \bullet T_F} \right] + 1$$
$$= int \left[ \frac{189,000}{13.5 \bullet 8.8 \bullet 144} \right] + 1 = 12$$

Initial cost of the membrane modules ( $C_{MM}$ ):

$$C_{MM} = C_{PM} \cdot N_{MM}$$
  
= 4,000 · 12 = 48,000

Total capital costs (C<sub>TC</sub>):

$$C_{TC} = C_{NM} + C_{MM}$$
  
= 17,000+48,000=65,000

Energy costs (C<sub>TE</sub>):

$$C_{TE} = \frac{E_{FS} + E_{CP} + E_{AC}}{\eta} \cdot C_{KW}$$
$$= \frac{7 \cdot 144 \cdot 52 + 7 \cdot 3 \cdot 52 + 4 \cdot 144 \cdot 52}{0.75} \cdot 0.103 = 11,460$$

Chemical cost (C<sub>cc</sub>):

$$C_{cc} = D_c \cdot V_c \cdot M_W \cdot C_{Bc} \cdot N_c$$
  
= 0.5 \cdot 200 \cdot 0.04 \cdot 10 \cdot 52 = 2,080

Membrane replacement costs (C<sub>MR</sub>):

$$C_{MR} = \frac{C_{MM}}{T_{Life}} + C_{Parts} = \frac{48,000}{5} + 1,000 = 10,600$$

Labor costs (C<sub>Labor</sub>):

$$C_{\text{Labor}} = 70,000$$

Total annual operating costs ( $C_{TO}$ ) for two-phase flow microfiltration:

$$C_{TO} = C_{TE} + C_{CC} + C_{MR} + C_{Labor}$$
  
= 11,460+2,080+10,600+70,000=94,140

#### VITA

Wen-Chang (Wayne) Liao was born in Taipei, Taiwan, on October 10, 1965. He attended National Cheng Kung University, Taiwan, and received a Bachelor of Science degree in Chemical Engineering in June, 1989. After spent two years on military service, he accepted a process engineer position with a polymer manufacturing company in Taiwan. In January, 1993, he accepted a teaching assistantship with the University of Mississippi, Oxford, Mississippi, Chemical Engineering Department and received a Master of Science degree in May, 1995. He attended the University of Tennessee, Knoxville, Tennessee, then accepted a position as Research Associate with the Food Science and Technology Department, while pursuing a Doctor of Philosophy degree in Biosystems Engineering with a concentration in Food Engineering.

He is a member of American Institute of Chemical Engineers and the Institute of Food Technologists. He is also a member of the Phi Kappa Phi and Gamma Sigma Delta honor societies.



