# ANALYZING THE EFFECT OF CROSS FLOW VELOCITY, UNIFORM TRANS MEMBRANE PRESSURE AND pH ON PERMEATE FLUX, RETENTATE COMPOSITION AND ENERGY CONSUMPTION DURING CROSS FLOW MICROFILTRATION OF SKIM MILK

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Mayank Singh

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## ABSTRACT

Pasteurized skim milk at pH 6.50 and 6.00 was microfiltered at 50$^{o}$C using 0.2-μm membranes. A factorial design of three cross flow velocities (CFV) of 5.3, 5.8, 6.3 ms$^{-1}$ and three uniform trans membrane pressures (UTMP) of 68.9, 103.4, 137.9 kPa was utilized. High CFV combined with high TMP required the shortest time (up to 40%) to achieve a concentration factor (CF) of 8-10x. Starting flux was 20 to 50% higher at pH 6.50 when compared to pH 6.00 due to solubilization of micellar calcium and severe fouling at lower pH. Higher flux was always obtained by using the combination of high CFV and high UTMP, which results in high shear and 33% reduction in gel layer ($C_G$) with back transport of rejected molecules into the retentate stream due to better (almost double) mass transfer coefficient ($k_c$). Higher CFV also reduced whey protein retention by 33%. Cross flow microfiltration (CFM) at lower pH (6.00) reduced calcium retention and lowered calcium to true protein ratio by 50% at 10x, compared to 8x retentate at pH 6.50, though 10x retentate had 20% higher casein concentration. Higher UTMP helped maintain high flux and thus shorten the run time up to a CF of 6-7x, but resulted in severe fouling and a steep decline in flux and increased whey protein retention as the process was continued to higher concentrations (8-10x). Overall energy consumption was always lower due to shorter CFM process when skim milk was microfiltered to 8x at higher CFV and higher UTMP.

## BIOGRAPHICAL SKETCH

In the spring of 1977, Mayank Singh was born as an elder son to Mr. O. P. Singh and Mrs. Sudershan Raghav in the holy state of Uttar Pradesh, India. He grew up in Roorkee, UA, the site of world's oldest engineering college east of river Nile, now known as I.I.T., Roorkee. Roorkee is a beautiful small town located in the Himalayan foothills on the banks of river Ganges, about 100 miles north of New Delhi. He finished his high school at St. Gabriel's Academy, Roorkee in 1994 with Mathematics and Science as majors. After getting admitted to National Dairy Research Institute in Karnal, India, he finished with an undergraduate degree of B. Tech. in Dairy Technology. Evaluating his options after graduation, Mayank had no hesitation in accepting offer for Master in Science program thousands of miles away at beautiful and prestigious Cornell University, NY working in Food Science and Engineering under the supervision of Prof. S. S. H. Rizvi. Since then, Mayank has been working in R&D division of Leprino Foods Company; world's largest natural cheese manufacturer, as a Senior Research Scientist, located in the heart of Rocky Mountains, Denver, Colorado. Mayank is married to Aditi Chauhan, who herself is working on her M.S. in Computer Science at University of Colorado and his younger brother, Shashank Singh is currently serving Indian Army as a Captain. Mayank is an avid bicyclist who also enjoys swimming, hiking, likes to play tennis and goes into hibernation from September to February when he enthusiastically follows Denver Broncos and the NFL.

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## LIST OF ABBREVIATIONS

| | |
|---|---|
| **CFM** | Cross Flow Microfiltration |
| **CFV** | Cross Flow Velocity |
| **CF** | Concentration Factor |
| **TS** | Total Solids |
| **TN** | Total Nitrogen |
| **CN** | Casein Nitrogen |
| **WP** | Whey Protein |
| **PF** | Permeate Flux |
| **KMH** | $kg.m^{-2}.h^{-1}$ |
| **UTMP** | Uniform Trans Membrane Pressure |
| **P** | Power |
| **P/F** | Power to Filtrate Ratio |

## LIST OF SYMBOLS

| | |
|---|---|
| $\gamma$ | Shear Rate ($s^{-1}$) |
| $Q_{ave}$ | Total volumetric flow rate through each membrane ($m^3.s^{-1}$) |
| R | Radius of each channel (m) |
| D | Diameter of each channel (m) |
| L | Length of the membrane (m) |
| m | Number of channels in each membrane |
| n | Flow behavior index |
| K | Consistency index ($Pa.s^n$) |
| $\tau$ | Shear stress (Pa) |
| J | Volumetric permeate flux (KMH) |
| dP | Trans membrane pressure (Uniform TMP/TMP) (kPa) |
| $\Delta P$ | Longitudinal pressure drop across the membrane (kPa) |
| $\mu$ | Feed viscosity (mPa.s) |
| v | Cross flow velocity from the flow meter (CFV) (m/s) |
| Re | Reynolds number |
| $\rho$ | Density ($kg.m^{-3}$) |
| f | Friction factor |
| $k_c$ | Mass transfer co-efficient ($kg.m^{-2}.h^{-1}$) |
| $C_G$ | Casein gel layer concentration (w/v) |

# CHAPTER ONE

# INTRODUCTION

## 1.1. Membrane Systems

Membrane based separation systems are well suited for liquid process streams and have found extensive uses in the food, pharmaceutical and biotechnology industries. Membrane filters have been commercially available since 1927 in Germany from the Sartorius Company, and since the 1960's, ultrafiltration had found its way into the dairy, pharmaceutical, biotechnology and chemical industries. By 1980s, a whole new range of value added dairy protein fractions were being manufactured made possible with the use of microfiltration. Among the food industries, dairy applications probably account for the largest share of installed membrane capacity (Cheryan, 1998). Today, ultrafiltration is widely used for fractionation of cheese whey and pre-concentration of cheese milk. Microfiltration's applicability in diary industry includes whey clarification, fractionation of casein micelles, whey proteins, milk fat, as well as concentration of unicellular materials of biological origin such as somatic cells, bacteria and other microorganisms. Casein enrichment of cheese milk to make hard/ semi-hard cheeses by MF is expanding rapidly because it significantly improves rennet coagulability. Cheese curd made with MF retentate is firmer and consequently leads to fewer fines in whey, which results in better cheese yields (IDF Bulletin, 1997). Research is continuing to understand many aspects of this intriguing science and technology.

Membrane science is the study of membrane development, improving the separation characteristics of existing membranes by making them less susceptible to harsh operating conditions, understanding the chemistry of the membrane and identifying

and using the optimum operating conditions to achieve maximum qualitative fractionation of desired components. Development of novel membranes can be achieved by carefully understanding the problems associated with membranes. Figures 1and 2 show the classification of various separation processes based on particle or molecular size. There are three major membrane separation processes for liquids commonly used in dairy industry:

1. Microfiltration (MF)
2. Ultrafiltration (UF)
3. Nanofiltration (NF)

These three membrane separation techniques are distinguished by the range of hydraulic pressure used to effect separation. However, it is important to note that although pressure is the driving force, it is the nature of the membrane which controls the movement of molecules through it. NF is a technique which has been classified in between Reverse Osmosis and UF. Its principal application is in the separation of mineral ions having a size in the nanometer size range (0.001-0.01μm). The industrial usefulness of this technique is dependent on the availability of special membranes, which are capable of separating mineral ions on the basis of their charge. UF can be viewed as a method for concentrating, fractionating and purifying fine colloidal suspensions such as cheese whey. UF retains only macromolecules (all nonfat milk solids except lactose and minerals), generally in the size range of (0.01-0.1μm), while microfiltration processes are designed to retain particles in range of 0.1-10μm. MF uses the most open or porous membranes in filtration spectrum. MF can allow skim milk to pass through and retain bacteria that are too large to pass through the membrane. The most promising and under-utilized MF technique is the separation of casein from serum milk proteins (whey). This application produces a casein-rich milk concentrate that is highly suitable for cheese making and a fat-free serum milk protein

stream that can be further processed with UF to make a whey protein concentrate. Typical MF systems used for milk processing will fully retain milk fat, casein and allow permeation of whey proteins, milk sugars and minerals. This technique provides a non-thermal processing alternative manufacturing for casein rich stream compared to the more traditional rennet and acid coagulation operations for casein separation. In addition, MF separated casein and whey proteins retain their native functionalities and are thus an excellent source of raw material for many dairy products. MF could also be used to manufacture high quality protein and protein concentrates without chemical usage with significantly reduced environmental impact (IDF Bulletin, 1997).

## 1.2. Fouling

Fouling during membrane separation leads to gradual decline of permeate flux and could affect the separation efficiency of a membrane due to pore blocking, pore bridging etc. A "fouled" membrane would not function properly to separate feed components on the basis of their size. Along with a decline in permeate flux, fouling is often accompanied by an increase in solute rejection; thus, a combination of these two factors have a direct impact on the permeation and mass transport properties of membranes and collectively constitute fouling. The initial rapid decline in permeate flux is primarily due to a phenomenon known as concentration polarization. During UF and MF, when the wall is porous, solids in the feed are brought to the membrane surface by convective transport, and a portion of solvent is removed from the fluid. This results in a higher local concentration of the rejected solute at the membrane surface as compared to the bulk feed. This solute buildup is also known as "concentration polarization". Depending on the type of feed, this layer could be fairly viscous or gelatinous. Thus a further resistance to the flow of permeate is encountered, in addition to the membrane and boundary layer. Concentration polarization builds up

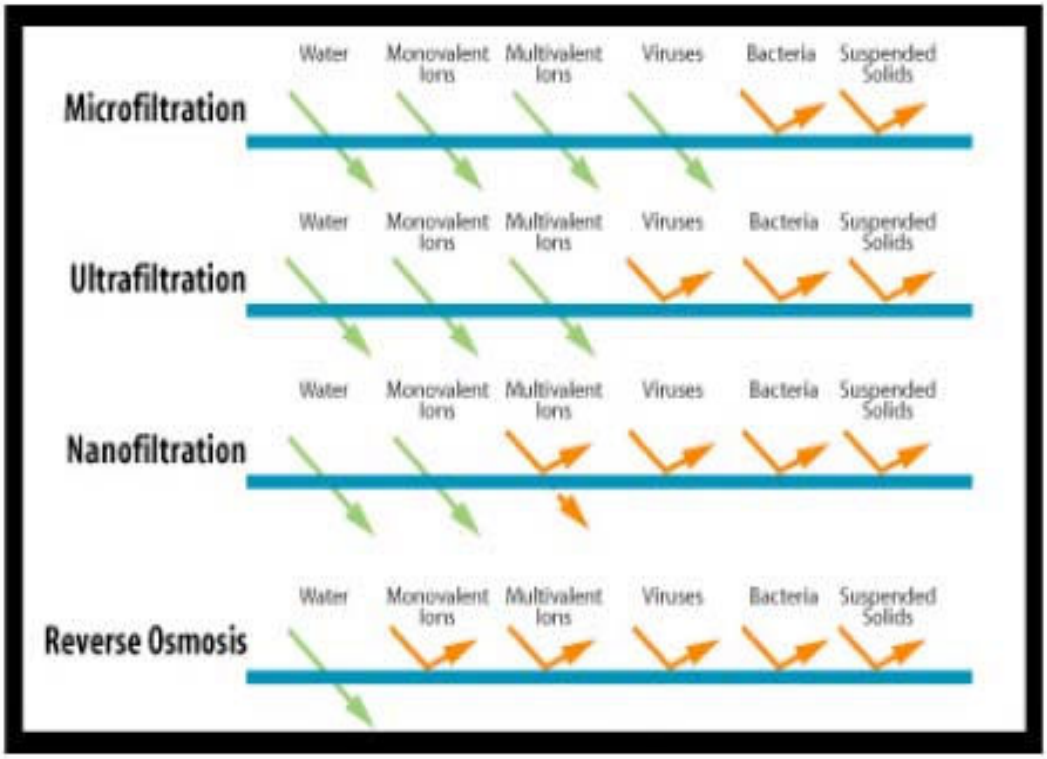


**Figure 1 Types of Pressure driven membrane processes**

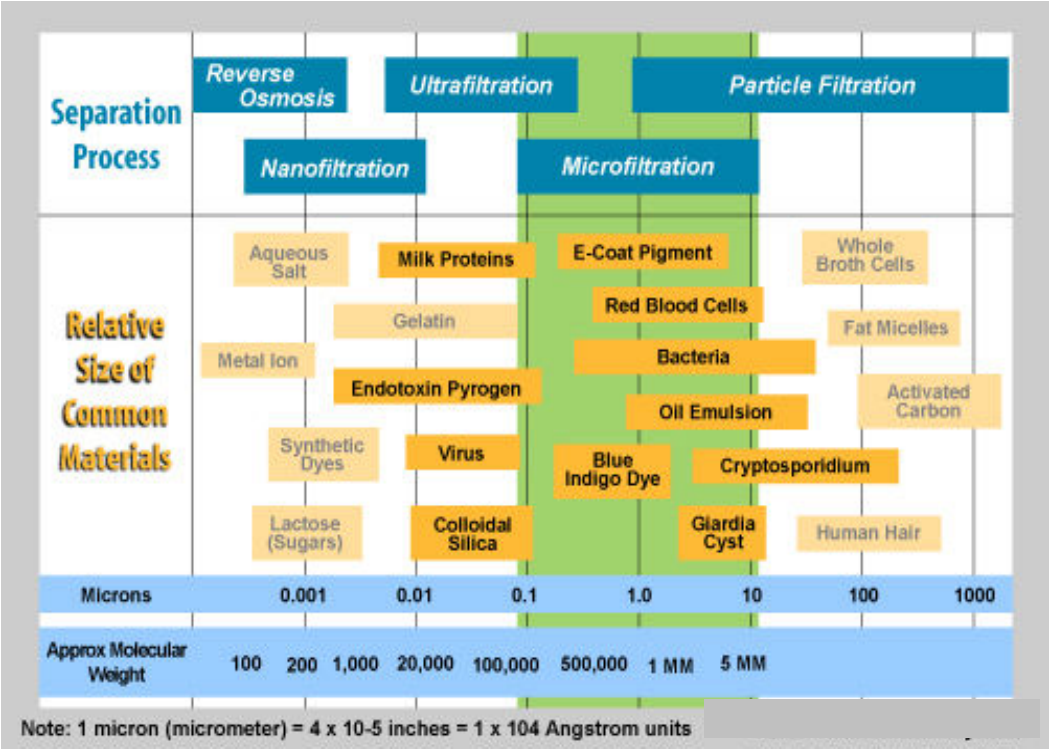


**Figure 2 Pressure driven membrane processes and their separation characteristics**

rapidly and results in lower mass transfer at the membrane surface. However, loss of flux due to concentration polarization is reversible and lost flux can be retained simply by flushing with water (IDF Bulletin, 1995).

The phenomenon that immediately follows concentration polarization is membrane fouling. Technically, membrane fouling is due to blocking of membrane pores by feed components, which as a result of various interactions get permanently deposited on the membrane and cause an increase in the concentration on the membrane surface. Fouling is mainly characterized by adsorption and pore blocking (Merin & Daufin, 1990). The various mechanisms involved in pore blocking depend on the size of the adsorbing molecule, surface reactivity of the membrane and pore size. Based on these factors, pore blockage can occur in the following fashion:

(i) Complete pore blocking

(ii) Pore bridging, which refers to the partial obstruction of the entrance?

(iii) Internal pore binding

Internal pore blocking is a serious fouling mechanism because bound molecules are not exposed to the effects of cross flow conditions. Howell and Nystrom (1993) classified proteins fouling typically seen in the dairy industry, according to two phenomena: *adsorption* and *aggregation*.

*Adsorption* refers to the chemical interaction between the membrane and the protein, which can be attributed to the high affinity of protein molecules for specific sites on the membrane surface. This stems from their relatively large size making multiple binding possible and the heterogeneity of the amino acid residues which comprise the protein, allowing it to form a variety of bonds: hydrophobic, van der Waals and polar.

*Aggregation* describes molecular association of proteins near the membrane surface forming entities of higher molecular weight. It is a two step process involving

denaturation and subsequent aggregation. Denaturation here refers to a change in the three dimensional structure of the protein from its native state and is a result of adsorption, change in pH, presence of salts, shear due to turbulent flow or temperature. Minerals and salts can have a profound effect on the fouling of membranes. On the one hand they can interact directly with the membrane as in the case of divalent ions where one of the positive charges interacts with the negatively charged sites on the membrane, leaving one positive charge free for interacting with negatively charged feed constituents. Divalent cations can have strong interactions with polysulfone membranes which are widely used in the dairy industry. Kuo and Cheryan (1983) suggested that an important way of controlling fouling is by adjusting the feed pH, which in turn controls the solubility of salts. Hayes et al. (1974) noted that the addition of calcium to cheese whey reduced the flux at pH 6.20. Patocka and Jelen (1987) found that eliminating calcium in cottage cheese whey increased the permeate flux. Vetier (1988) performed ultrafiltration trials with various calcium contents and concluded that the most important role of calcium is that it acts as a binding agent between the protein layers.

### 1.3. Milk Chemistry

Milk may be defined as the whole, fresh, clean, lacteal secretion obtained by the complete milking of one or more healthy milch animals, excluding that obtained within 15 days before or 5 days after calving or such periods as may be necessary to render milk practically colostrum-free (De, 1980). Chemically, skim milk can be classified as a lyophilic colloid, because the protein complexes of skim milk, which constitute the dispersed phase, are in the correct size range to interact with and are stabilized by the solvent and do not spontaneously, coagulate. The milk protein

colloidal solution is stable to gravitational force, but can be separated by centrifugation.

The major components of skim milk are casein, whey proteins, sugars, minerals and vitamins.

**CASEIN:** It constitutes about 80% of total milk protein and has three distinct fractions: α, β and γ-casein. However, a casein sample from pooled milk when subjected to gel electrophoresis yields up to 20 casein components in the size range of 25 to 300 nm (Table 1). Casein precipitates out at pH 4.60 and 20$^{o}$ C. In normal milk, approximately 95% of casein exists as micelles, with mean diameters of about 100 nm. Based on their size, it is expected that all casein protein will be retained during MF. Micelles are made up of casein submicelles and micellar calcium phosphate. It is generally accepted that κ-casein is found at the micelle surface and is in part, responsible for micelle stability. During proteolysis by rennet κ-casein is acted on and removed from casein macro-peptide, which destabilizes the micelle and the caseins aggregate and gel in the presence of calcium. Micelle stability is also affected by temperature, pH and calcium content. As the pH decreases, more calcium and phosphate dissolve and are removed from the micelle. All the calcium phosphate is soluble below pH 4.90.

***α-Casein*** has further been classified into α-$_{s1}$, $\alpha_{s2}$, $\alpha_{s3}$ etc. $\alpha_{s1}$ casein is a single chain polypeptide of known sequence of 199 amino acid residues, and constitute 50% of total casein. This component shows a high degree of hydrophobicity responsible for the pronounced self-association of the $\alpha_{s1}$ casein monomer in aqueous solutions, approaching a limiting size under most conditions of ionic strength.

***β-Casein,*** the second most abundant milk protein (35% of casein) is a single chain with 5 phospho-serine residues. In aqueous solutions, β-casein undergoes an endothermic self-association, which reaches a maximum or limiting size, depending

upon the ionic strength of the solvent. β-Casein is much more "soap like" than $\alpha_{s1}$casein.

***κ-Casein*** is third most important milk protein (12% of casein) and is soluble over a very broad range of calcium ion concentrations and performs an important role of casein micelle stabilization. κ-Casein also shows soap like properties. The C-terminal of the molecule, which constitutes 1/4th of the structure, is quite hydrophilic and accounts for all the net charge of the κ-casein molecule.

**WHEY PROTEIN:** The serum or whey proteins are those which remain in solution after the casein are removed, and these proteins are not incorporated in colloidal complexes. Their size varies from 3 to 5 nm. Depending upon the extent of denaturation and method of separation, different amounts of whey proteins are included in the retentate during MF.

**α-lactalbumin** is the smallest of whey proteins and accounts for 25% of total whey proteins. α-lactalbumin is very stable with 4 disulfide cross links, has a spherical shape and is known to bind calcium.

**β-lactoglobulin** are typical globular protein and account for 60% of whey proteins. They exist as a dimer at room temperature between pH 7.00 and 5.20 and dissociates into monomers at temperatures above $40^{o}$ C. Conditions such as excessive heat or low pH causes irreversible denaturation and coagulation of β-lactoglobulin.

**Other serum proteins:** Serum albumin and immunoglobulins occur in skim milk to limited extent, in conjunction with various enzymes. In fact, protein denaturation also involves, in part, denaturation of many of these enzymes. However, these proteins do not have a significant impact on fouling.

**Role of calcium:** Kessler et al (1982) investigated the effect of adding lactose and calcium to an aqueous protein suspension containing 3.5% protein. Adding lactose had little effect on UF flux and hence the protein deposit. In contrast, adding calcium

resulted in a considerable decrease in permeate flux, approaching that achieved with skim milk. Vetier et al (1988) experimented with milks of various calcium contents using a 0.2 μm ceramic MF membrane. Fouling increased with increasing calcium content. In cases where calcium levels were reduced in whey, permeate flux was observed to increase.

**Table 1. Milk Components and their size**

| *Component* | *Size (nm)* |
|---|---|
| Water | 0.3 |
| $Ca^{2+}$ | 0.4 |
| Lactose | 0.8 |
| Whey Proteins | 3 – 5 |
| Casein micelles | 25 – 300 |
| Fat globules | 100 – 2000 |
| Bacteria | 200 – 8000 |

Webb, B., Johnson A. and Alford, J. 1987. Fundamentals of Dairy Chemistry. CBS Publ., Delhi, India

## 1.4. Microfiltration of Milk

Cross-flow microfiltration (CFM) is a pressure driven membrane process, which could be used for coarse filtration of particulates and bacteria, as well as for fine fractionation of proteins, small molecular weight solutes and water. Due to the nature of milk constituents, there is a tendency for large particles or colloidal aggregates to be trapped in the pores followed by a cake formation on the surface of membrane and creation of new dynamic resistance layers. This layer starts to govern the overall filtration characteristics and is independent in its rejection properties of the initial pore size of the membrane. Influence of TMP was found to be a critical parameter of flux decline during CFM of skim milk (Maubois, 1998). It was shown that when TMP was increased, fouling occurred much faster. MF should therefore operate at high CFV in order to limit fouling. This results in high TMP due to high pressure drop along the filtration path and in an uneven distribution of fouling layer. In order to avoid this phenomenon, it was proposed to establish a uniform low TMP profile along the filtration path by implementing a patent registered by Alfa Laval Company (1974). It consists of circulating permeate co-current to the retentate, and provides uniform fouling of membrane surface, allowing milk to be concentrated multiple times and providing novel retentates with very high concentration of casein solids. The development of MF will facilitate commercialization of native micellar casein and "liquid virgin whey protein concentrate" (Korat and Rizvi, 2004). Both products in turn can be used as starting materials for the separation and purification of individual caseins and whey proteins. Another exciting area for future exploration is the isolation of biologically active peptides (Damodaran and Paraf, 1997). All of the milk proteins appear to contain peptide sequences possessing biological activity. Peptides with opioid activity, called exorphins, have been identified in most milk proteins, of which,

the most well known are β–casomorphins. This family of peptides, containing 4 to 7 amino acids with common N-terminal sequence, Tyr-Pro-Phe-Pro, are very resistant to enzymes of gastrointestinal tract and appear in the contents of small intestine following ingestion of milk and also in blood plasma of pregnant and lactating women and newborn infants. All these peptides have been shown to have opioid activities, which include regulation of electrolyte transfer, pain suppression and sleep induction. Caseins contain sequences called casein phosphopeptides that play a major role in bioavailability of calcium and iron. Portions of macro-peptides could help with blood pressure regulation and inhibit blood coagulation by blocking specific receptors on platelets.

Further work is obviously needed on the extraction and purification of many milk components. What this requires is a through understanding of the phyico-chemical properties of these components and development of sophisticated separation techniques. Work done in this research project is focused on employing CFV and UTMP and pH in order to economically achieve maximum fractionation of casein and whey proteins, while looking for suitability of retentate for cheese making. This knowledge could be further applied to further fractionate casein rich retentate and high quality whey streams.

## 1.5. Common Definitions

*Cross-flow Microfiltration (CFM)*: The fluid stream is passed tangentially to the membrane surface with a high cross flow velocity & some TMP.

*Concentration Factor (CF):* The ratio of initial feed volume (or weight or flow rate) to the retentate volume (or weight or flow rate) at any given time. For example, when a 200 kg feed has been reduced to 50 kg retentate during microfiltration, the CF would be 4x.

*Channel:* The space in membrane module where feed flows, e.g., the inside of hollow fibers or tubes

*Cross Flow Velocity (CFV):* The linear rate of flow of fluid parallel to the membrane, expressed in units of length/time (meters / second) It is calculated as flow rate / cross-sectional area of feed channel

*Delta (Δ) P:* Pressure drop, defined as inlet pressure minus outlet pressure

*Fouling*: A phenomena in which membrane absorbs or interacts in some manner with the feed components, resulting in a decrease in membrane performance.

*Permeate:* The portion of feed solution that passes through the membrane.

*Permeate Flux:* Amount of liquid passing through the membrane expressed in terms of volume through unit area in unit time, e.g., $kg.m^{-2}.h^{-1}$ (KMH)

*Retentate:* The portion of feed solution that is retained on the high pressure side of the membrane.

*Reynolds Number (Re):* A measure of state of turbulence in a fluid system. It is calculated as the ratio of inertia effects to viscous effects. Fluid flow with Re value less than 2100 is considered to be laminar.

*Trans Membrane Pressure (TMP):* The driving force for flux. In our cross-low microfiltration system, it is measured as the difference between retentate and permeate pressures at the inlet and outlet.

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# CHAPTER TWO

# EFFECT OF OPERATING PARAMETERS ON CROSS FLOW MICROFILTRATION PERFORMANCE ON SKIM MILK

## 2.1. ABSTRACT

Pasteurized skim milk (200kg) was microfiltered at 50$^{o}$C using 0.2-μm membranes at cross flow velocities (CFV) of 5.3, 5.8, 6.3 ms$^{-1}$ and uniform trans membrane pressures (UTMP) of 68.9, 103.4, 137.9 kPa at pH 6.50 or 6.00. As expected, increasing CFV at each UTMP improves the starting flux, with an average increase of 34% for only a 20% increase in CFV. High CFV combined with high TMP gave the highest initial flux and shortest time required to achieve concentration factors (CF) of 8x at pH 6.50 and 10x at pH 6.00, even though there was a sharp decline in permeate flux after CF of 6x. Increase in CFV lowered WP levels by 44% and 33% in final retentate at pH 6.50 and 6.00, respectively. Increasing CFV lowered calcium levels by 5% at pH 6.00. Increase in UTMP also increased total solid levels in retentate. Much of this increase was contributed by whey proteins, where up to 25% and 33% higher whey protein level was measured in final retentate at pH 6.50 and 6.00, respectively. In every experiment, flux at each CF was higher at pH 6.50 when compared to pH 6.00 due to solubilization of micellar calcium and severe fouling at lower pH.

## 2.2. INTRODUCTION

The development of robust ceramic membranes with multi channels in the 1980s led to the acceptance of cross flow microfiltration as a viable industrial separation technology remarkable enough to alter the very fabric of traditional dairying practices. Milk is a multi-component and multiphase system, and it's fractionation into value-

added components of unique physico-chemical characteristics offers the potential to optimize the production of value-added milk components and enhance their utilization in a variety of food formulations. The three most common uses of microfiltration in dairying have been: i) cold pasteurization (or removal of microorganisms), ii) concentration and clarification of whey, and iii) fractionating skim milk into casein-rich retentate and whey protein-rich permeate (Korat and Rizvi, 2004; Maubois, 2002; Brandsma and Rizvi, 1999).

Removal of bacteria using cross flow microfiltration has been widely studied, and bacterial retention of more than 99% has been reported, being 99.99% for species commonly found in raw milk (Trouve et al, 1991). Kelly & Tuohy (1997) have reported an average of 99.1% removal of mesophilic bacteria and 99.3% removal of themoduric bacteria.

CFM also has been used to clarify cheese whey by removing lipids, colloids, bacteria and other aggregates, which can be further ultrafiltered for substantially improved flux (Maubios, 2002).

Depending on the membrane pore size (0.1-10 μm), microfiltration can be used to concentrate macromolecular milk constituents like casein, fat globules, WP aggregates, cheese fines and bacteria. The most promising application of CFM seems to be the selective concentration of casein micelles in their native state. This technology not only produces permeate with chemical composition and quality superior to sweet whey, but also native casein enriched retentate, highly desirable as is (Maubios, 2002; Pouliot and Pouliot, 1996) and for vat-less cheese manufacture (Korat and Rizvi, 2004). Significance of CFM to dairy industry lies in improving cheese making, increasing yield, and enhancing the value of by-product ("ideal whey") (Korat and Rizvi, 2004; Brandsma & Rizvi, 2001). During microfiltration (0.2μm), fat globules, casein and immunoglobulins are completely retained and,

fractions of α-lacta albumin, β-lacto globulin, lactose, minerals and vitamins are partially retained, mostly re-distributed with water phase. The diameter of WP is 3-5 nm, and that of casein micelles is 25-300nm (Walstra and Jenness, 1984). As the retention of proteins steadily increases with time and after a substantial pore plugging, MF starts behaving as UF.

A remarkable difference in UF and MF is the partial retention of WP by MF membranes. On a molecular level, UF fouling is for most part on the membrane surface, whereas severe fouling occurs in MF due to pore plugging (Marshall et al. 1997). The ultimate endeavor of this research project was to make highly concentrated retentate in the shortest possible time with minimum retention of WP. While UF retentates include all WP leading to functional defects in cheese made from it, the cheese made from MF retentates have less than 50% of WP, and this brings dramatic improvement in functional properties still providing improved cheese yields due to incorporation of some WP (Brandsma & Rizvi, 2001).

The membrane separation of complex biological protein solutions such as milk is accompanied by a progressive increase in fouling leading to a gradual decline in permeate flux. From the work done so far, it is understood that although membrane fouling mechanism is predominantly a function of the experimental conditions, it is also affected by membrane characteristics and feed properties. Several studies have been done to evaluate the CFM process in terms of permeate flux. Sachdeva & Buchheim (1997) studied the effect of initial permeate flow rate (which is proportional to TMP applied) on the flux decline during CFM (0.1 μm) of skim milk up to CF= 4. However, TMP was changed throughout the experiment (0.7-1.1 bar) for minimum decline in flux, so permeate flux under constant TMP was not measured. Instead, effect of permeate flux on TMP was studied. Also, they did not study the effect of

CFV. Their study reported very rapid flux decline and higher retention of WP at higher initial flux rates.

Berre & Daufin (1996) studied the CFM performance (0.1µm) with respect to permeate flux to wall shear stress ratio ($J/\acute{\imath}_{weff}$), rather than individually analyzing the effect of CFV and TMP. The $J/\acute{\imath}_{weff}$ ratio constitutes a basic parameter which characterizes competition between convection and erosion at the membrane / solution interface. They reported that operating over a critical value of $J/\acute{\imath}_{weff}$ (1.01) resulted in lower WP retention with slower fouling. However, their study evaluated protein retention at very low CF (2x), and the effect of TMP was not determined because TMP was changed throughout the experiment. More than 99% casein retention was reported in all cases, and higher shear rates caused an increase in WP retention, pointing out that CFM may not have to be performed at the highest attainable CFV.

Some other studies with MF membranes have shown that higher CFV and TMP result in higher permeate flux and lower membrane fouling (Vyas et al. 2002; Samuelsson et al., 1997). Samuelsson et al. (1997) also studied the effect of operating conditions on permeate flux and WP retention. However, their system was vastly different from the experimental set up used in this study. They achieved a final CF of only 1.15, operating under non-UTMP mode, and they did not include any details on the chemical analysis of retentate.

Colloidal fouling of MF membranes (0.2, 1, 5, 10µm) using calcite and anatase suspensions was studied by Wakeman & Traleton (1991). In all cases, increased filtration pressure resulted in an improved filtration rate. For coarse calcite suspensions, increasing CFV had a detrimental effect on flux, attributed to faster migration of larger particles from the membrane surface, leaving smaller particles for

fouling. These results were contradictory to most CFM studies done on milk such as Vyas et al (2000) and Samuelsson et al. (1997), who have suggested to increase shear rate on membrane surface in order to sweep away the accumulating particulates retained by the membrane.

Several microfiltration studies have been done on skim milk fractionation operating with a uniform permeate flux mode and/or non UTMP mode at low concentrations. In uniform permeate flux mode, permeate was removed at a constant rate, and to do this, trans membrane pressure was increased until it could not be increased any more, resulting in severe fouling. In some other cases, no permeate was re-circulated causing non-UTMP which leads to differential fouling across the length of the membrane resulting in under utilization of the membrane surface. Review of such studies and benefits of UTMP have been clearly established (Vadi and Rizvi, 2001). All experiments in this study were performed at UTMP.

There are no studies published to date that show the effect of pressure and velocity on the CFM performance and it's influence on permeate flux and the efficiency of separation, using UTMP to achieve high concentrations such as 10x. Limited research has been done on this concept by some researchers like Sachdeva and Buchheim (1997), Berre and Daufin (1996) and Samuelsson et al. (1997) but there is no work that combines the qualitative and quantitative performance of CFM. The objective of this study was to establish the effects of CFV and TMP on fractionation of skim milk proteins using a UTMP system.

## 2.3. MATERIALS AND METHODS

### 2.3.1 Microfiltration System

The cross flow microfiltration assembly (Figure 3.) used in this study had two 0.2-micron membrane elements (1P/R19-40, US Filter Corp., Warrendale, PA). Each membrane element had 19 channels, with a diameter of 4 mm. The length of each element was 850 mm, yielding a total surface area of 0.4 $m^2$. Both elements were placed in separate parallel housings. Later a second module consisting of three membrane elements of the same pore diameter but of greater length (1050 mm) and greater surface area (0.72 $m^2$) was added to enhance the filtration capacity of the system. The inlet and outlet pressures on both retentate and permeate sides were measured with pressure gauges (Anderson Instrument Co.), and could be controlled by flow control valves. The feed flow rate was kept constant with the help of flow meters (Series 55-200, Wallace & Tiernan, Belleville, NJ) attached at the inlets of both membrane elements. The pump used for retentate circulation was centrifugal and water-cooled (7.5HP, Reliance Electric Co.) and the permeate circulation pump was centrifugal and air-cooled (5 HP, Reliance Electric Co.). A coaxial heat exchanger was used to maintain the retentate temperature during it's return to the feed balance tank. In addition, a constant head was maintained on permeate side with a small overhead tank (12 kg), and permeate flow rate was adjusted to achieve uniform trans membrane pressure. Any net overflow was measured as the flux of the system. The system was operated in batches, and the dead volume of the system was 16 kg.

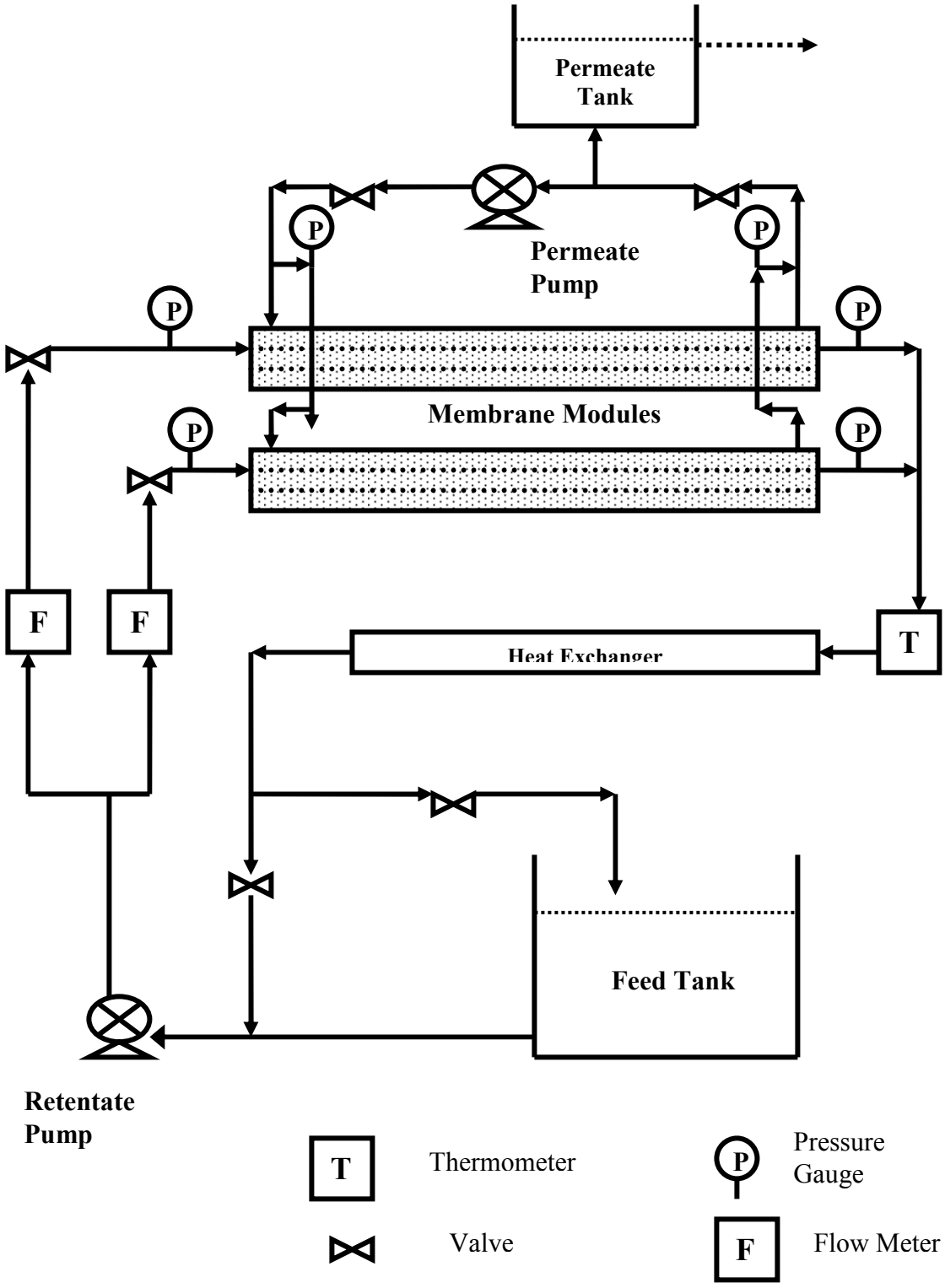


**Figure 3. Schematic of Cross Flow Microfiltration System**

### 2.3.2. CFM Process

Pasteurized skim milk was microfiltered at 50$^{o}$C using CFM (0.2-µm pore size) at three CFVs (5.3, 5.8 and 6.3ms$^{-1}$) and three UTMPs (68.9, 103.4 and 137.9kPa) in duplicate runs in a two-level 3x3 factorial design at pH 6.50 and 6.00. Volumetric CF of 8x was achieved at each combination of CFV and UTMP at pH 6.50. Pressure was measured in "psi" through pressure gauges and later converted into kPa for calculations. Pressure drop across the membrane length was compensated by adjusting the permeate flow to achieve uniform trans membrane pressure throughout. For example at CF = 4, the retentate/permeate pressures were 448.2/310.3 kPa at the inlet and 337.9/200.0 kPa at the outlet, giving a UTMP of 137.9 kPa and a longitudinal pressure drop of 110.3 kPa. Similarly at pH 6.00, CFM was carried out at the same three CFVs (5.3, 5.8 and 6.3 ms$^{-1}$) and three UTMPs (68.9, 103.4 and 137.9kPa) in duplicates, and volumetric CF of 10x was achieved at each combination of CFV and UTMP. To lower the pH during CFM, a known amount of glucono-delta-lactone was added. Retentate temperature was maintained at 50 ± 1$^{o}$C by recirculating cold water at 4$^{o}$C. The permeate flux expressed in KMH (kg-m$^{-2}$-h$^{-1}$) was measured every 10 min, and the pressures and flow rates were recorded. Samples of retentate and permeate were collected and analyzed for chemical composition.

### 2.3.3. Cleaning

After the end of each experiment, washing fouled membranes with cleaning solutions restored clean water flux. After rinsing with regular water, first a 2% w/w NaOH solution at 70 $^{0}$C was circulated for 1.5 hrs without opening the permeate line. Then the permeate flow valve was opened to clean any plugged membrane pores, and the solution was circulated for another 1.5 hrs though the membrane pores. In the end, the

hot alkali solution was pushed to the permeate tank, and reverse circulation was done for 45 min. These three cycles were also repeated for 2% $HNO_3$ w/w at $70^o$ C after rinsing the alkali out. Finally, the membrane was rinsed and the water flux at 137.9 kPa (9000 KMH) was checked before the start of next experiment. During cleaning, the temperature was increased or decreased 1 $^o$ C / min to prevent heat shock to the membranes.

### 2.3.4. Statistics

MINITAB release 9 (Minitab Inc., State College, PA) and Microsoft Excel (Microsoft Corp, Redmond, WA) were used for statistical analysis of the data. Permeate flux during CFM of skim milk was measured every 10 min. The effect of CFV was assessed by using 3 different CFV (5.3, 5.8, 6.3 $ms^{-1}$) at fixed TMP using ANOVA. Similarly, the effect of TMP was quantified by using 3 different TMP (68.9, 103.4, 137.9 kPa) at a given CFV using ANOVA. Significant differences were determined at $P < 0.05$. The whole experiment was formulated into a 3x3 factorial design at each pH. Experiments were performed in random order to negate any carry over effects. After every run was completed, the permeate flux as a function of CF was plotted. A best – fit (polynomial) regression line was drawn through the data set and the instantaneous flux values at each CF were calculated from the graph. This exercise was repeated for all the runs to get flux values at each CF. All samples were analyzed for chemical composition in duplicates. Analysis of variance was used to determine the effect of CFV and UTMP on major component retention.

### 2.3.5. Compositional Analysis

Total solids were determined using forced oven drying (AOAC, 1995) and fat by Mojonnier ether extraction (AOAC, 1995). Ash was determined by drying samples in

a forced air oven at 100$^{o}$C and then placing the sample dish in a muffle furnace for 20 h at 550$^{o}$C. Nitrogen content (total nitrogen (TN), non-casein nitrogen (NCN), NPN) in samples was determined by Kjeldahl method (AOAC, 1995). Total protein was calculated by multiplying the total nitrogen by 6.38. True protein was calculated as (TN - NPN) x 6.38 and casein as (TN - NCN) x 6.38. Whey Protein (WP) was calculated as (NCN – NPN) x 6.38. Total calcium was measured by atomic absorption spectroscopy analysis, as described by Metzger et al (2000). Finally, lactose was calculated by difference. All samples were analyzed in duplicates.

## 2.4. RESULTS AND DISCUSSIOIN

### 2.4.1. Effect of CFV on Permeate Flux

Increasing the CFV at constant UTMP resulted in up to 34% improvement in starting permeate flux at pH 6.50 and 60% improvement at pH 6.00. Figures 4 and 5 show the effect of CFV on permeate flux at various CF and different pH. Tables 7 and 10 (in *Appendix*) show the exact time needed to reach a particular concentration factor at pH 6.50 and 6.00 respectively and Tables 8 and 11 (in *Appendix*) rank both sets of 9 runs based on time needed to reach a concentration factor. It was observed that using highest CFV and highest UTMP could reduce required CFM time by 37 % to 42% (for 8 to 10x), when compared to lowest CFV and lowest UTMP used in this study. The difference achieved by increasing CFVs at any concentration factor or pH was always statistically significant ($P < 0.05$), except between CF 5x to 6x at pH 6.00.

The initial flux increased by about 34% with 20% increase in CFV, reflecting improved mass transfer of rejected solutes back to the bulk solution and prevented fouling and gel layer concentration (Figures 6 and 8). This increase in flux is somewhat greater than expected, based on published correlations for mass transfer in

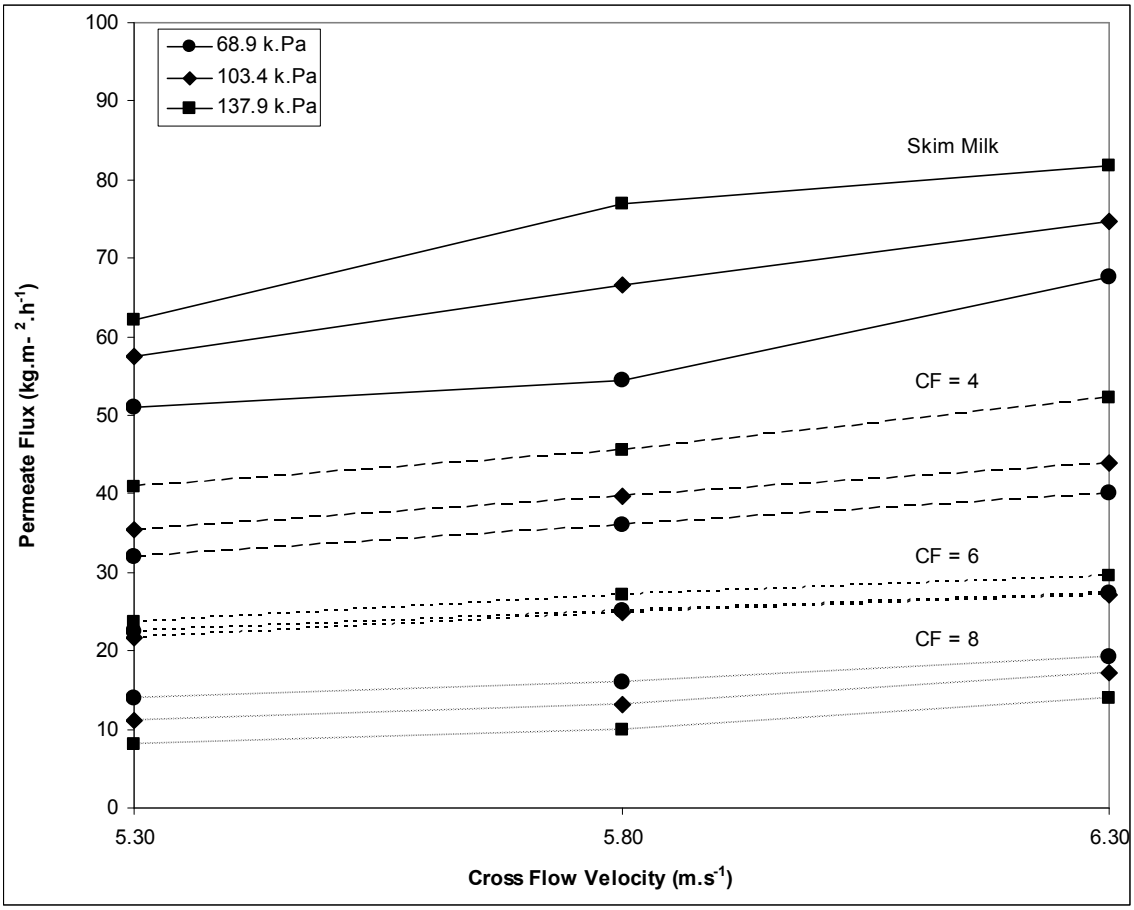


**Figure 4. Effect of CFV x UTMP on Permeate Flux at selected CF (1, 4, 6, 8) at pH 6.50**

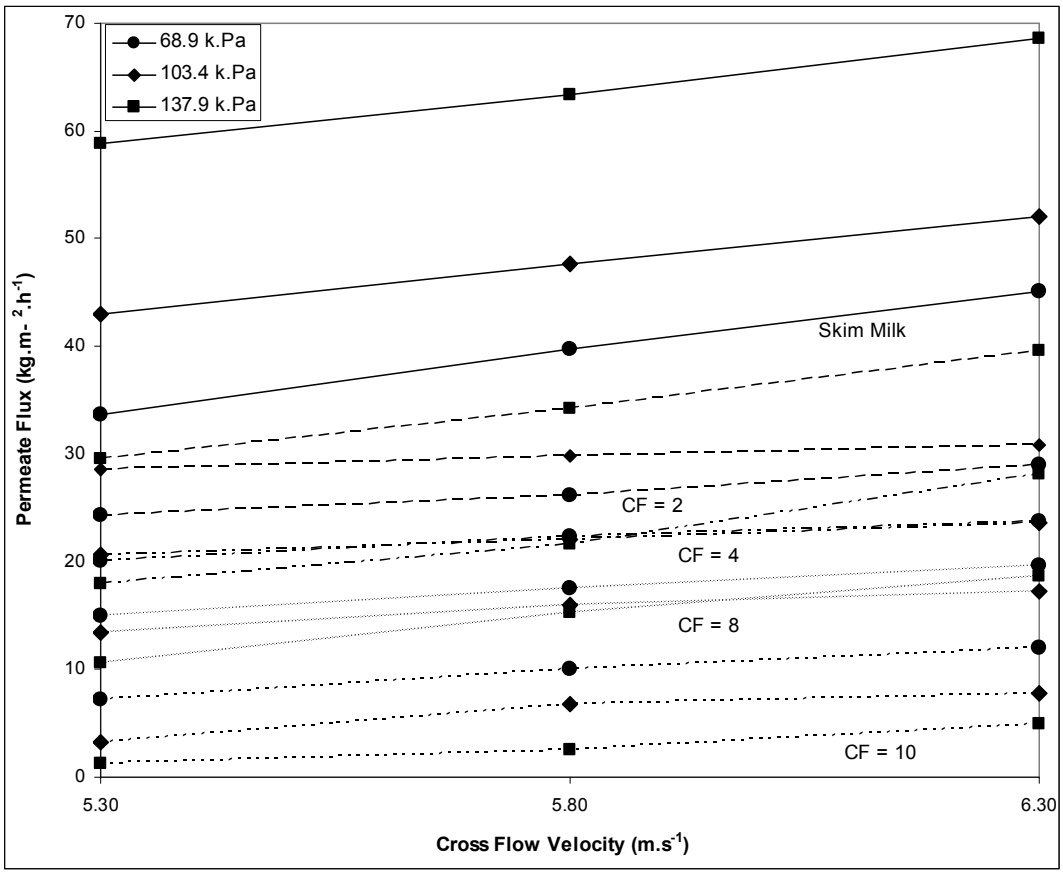


**Figure 5. Effect of CFV x UTMP on Permeate Flux at selected CF (1, 4, 8, 10) at pH 6.00**

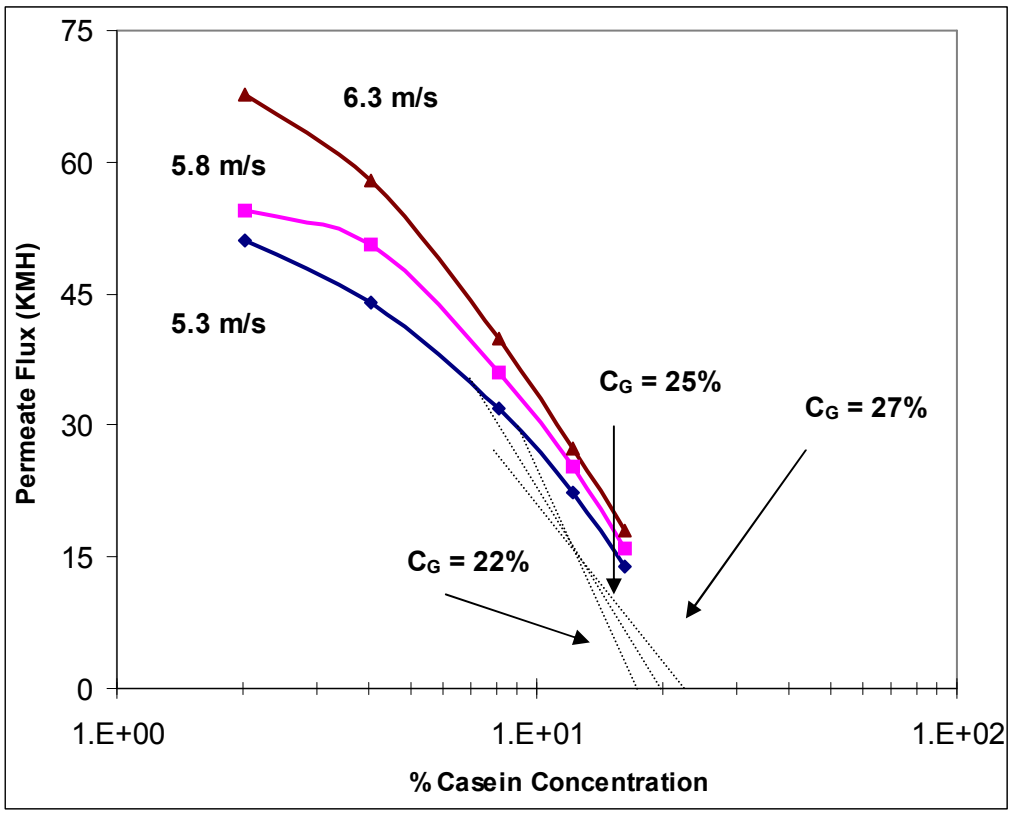


**Figure 6. Effect of CFV on casein gel layer concentration ($C_G$) at pH 6.50**

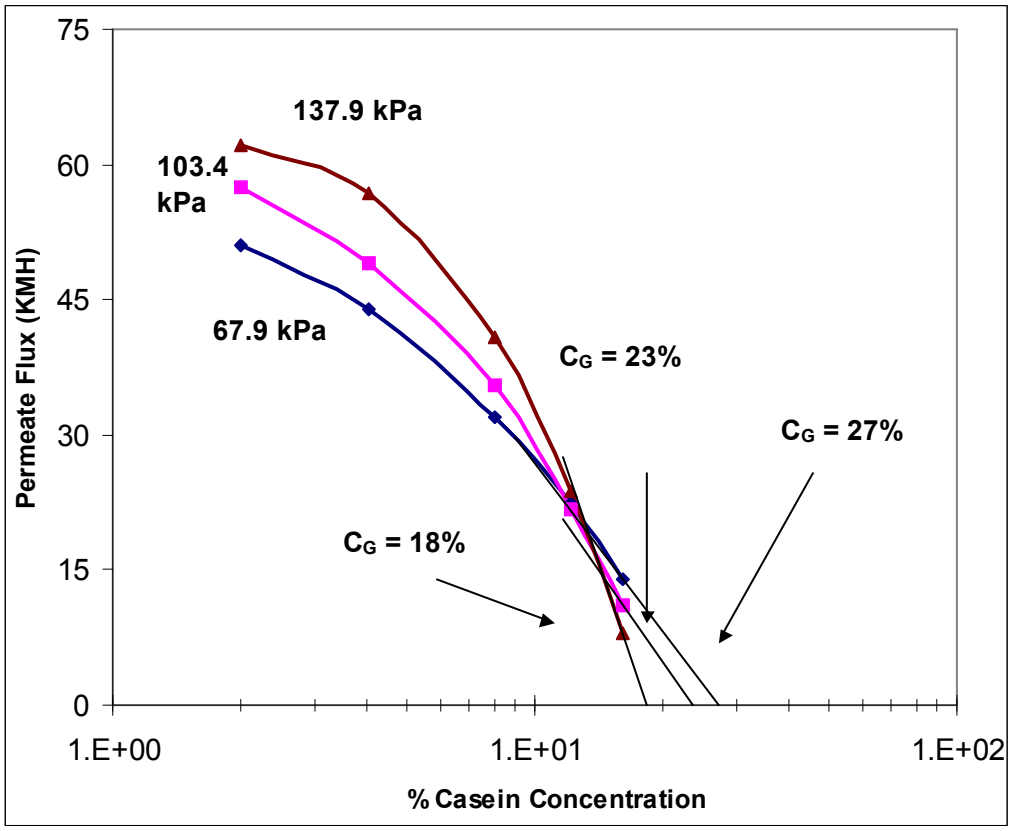


**Figure 7. Effect of UTMP on casein gel layer concentration ($C_G$) at pH 6.50**

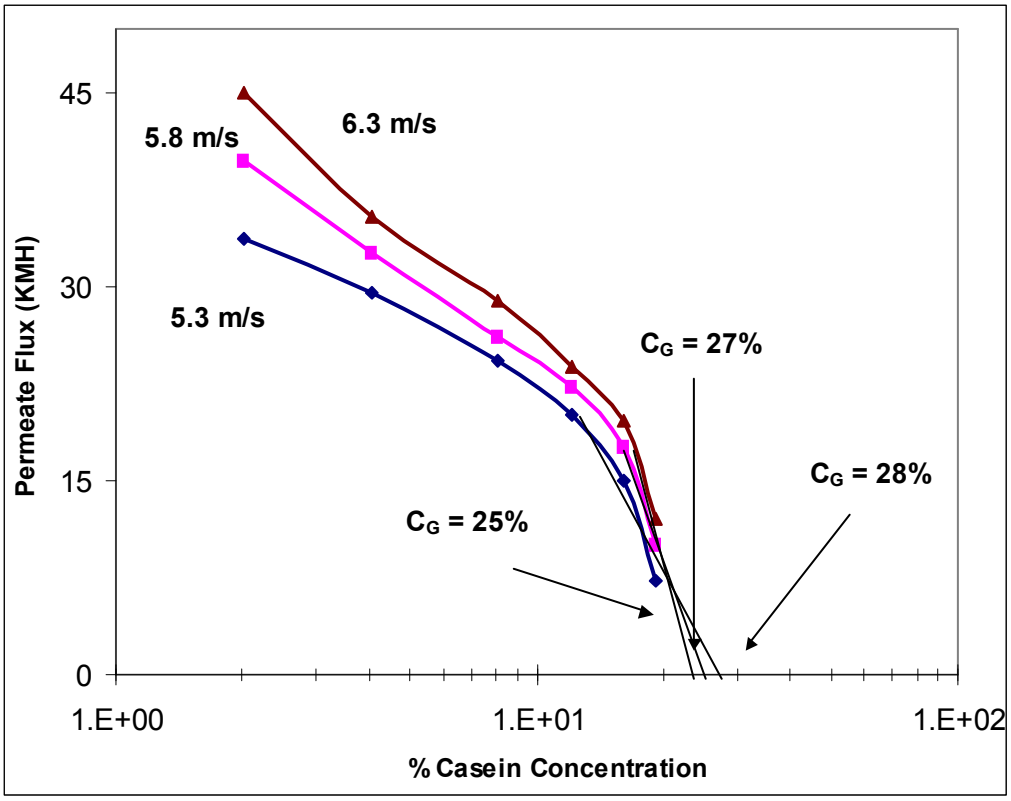


**Figure 8. Effect of CFV on casein gel layer concentration ($C_G$) at pH 6.00**

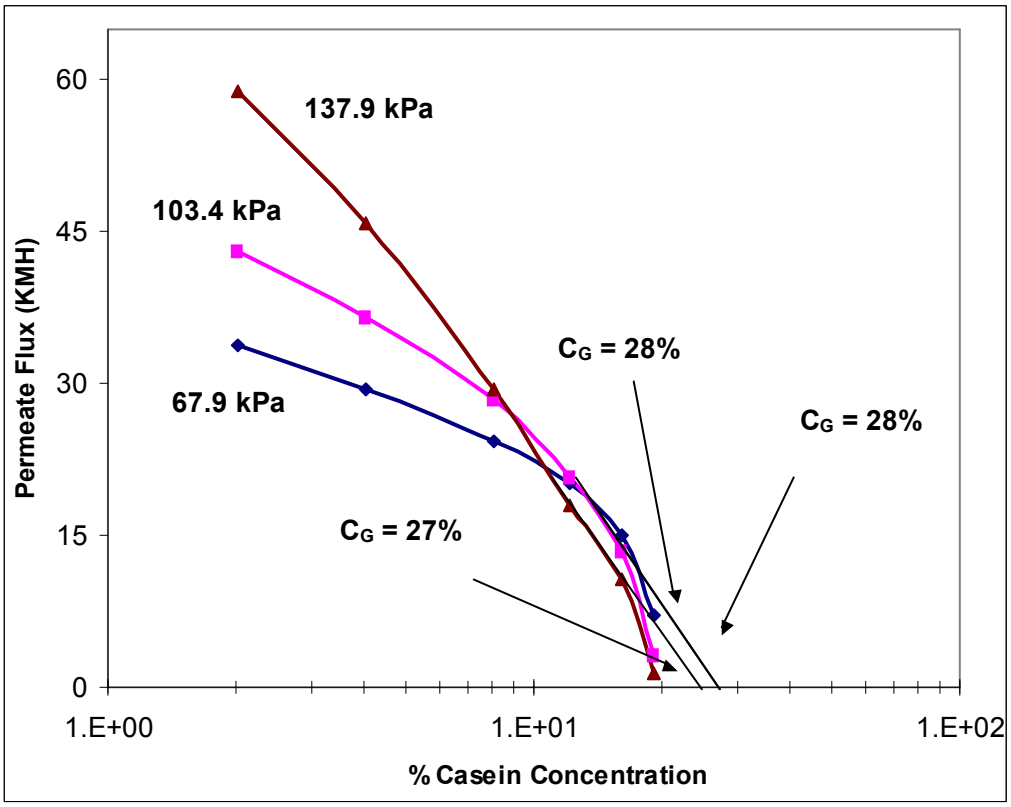


**Figure 9. Effect of UTMP on casein gel layer concentration ($C_G$) at pH 6.00**

turbulent flow, where the mass transfer coefficient varies with the 0.8 to 0.9 power of the flow rate. At CF of 4 or 6, the increase in flux with CFV was about 25 %, but at CF beyond 8x, it was as high as 75%. However at CF greater than 8x, the flux was so low due to membrane fouling that the direct effect of velocity or shear is hard to evaluate. The results follow the same trend at each CF and pH as shown in Figure 4 and 5. We did not collect sufficient data to estimate exact degree of fouling, but as expected, higher CFV was critical in getting significantly higher permeate flux rates at every CF.

Using a semi-log plot of casein concentration against permeate flux (Figure 6 and 8), it was estimated that increase in CFV at constant UTMP almost doubled the mass transfer coefficient ($k_c$) and reduced gel layer concentration ($C_G$) by up to 22% (from 27% to 21%). Other authors including Samuelsson et al. (1997) and Clarke & Heath (1997) have reported similar effects of CFV during ultrafiltration of skim milk. Their model correlation indicated that increase in velocity leads to an increase in mass transfer coefficient or a greater decrease in boundary layer resistance, which would result in an increase in permeate flux. Evidence of reduced membrane fouling during high velocity runs is given by the fact that whey protein retention decreases with increase in CFV, probably due to increase in effective pore size (Vyas et al. 2000; Marshall et al. 1997).

### 2.4.2. Effect of UTMP on Permeate Flux

Increase in UTMP at constant CFV resulted in a sharp decline of permeate flux against time at both pH as shown in Figures 4 and 5. When these values were used to estimate gel layer concentration (Figures 7 and 9), it was seen that a two-fold increase in UTMP could reduce the casein concentration in gel layer by up to 35%. At pH 6.50,

the starting permeate flux, (Figures 4 and 5) was as up to 22% higher when UTMP was doubled (from 68.9 kPa to 137.9 kPa). The small effect of UTMP is similar to the reports for ultrafiltration of skim milk at low to moderate TMPs (Cheryan, 1986), and it indicates a small but not negligible membrane resistance. At higher TMP, the flux may become independent of TMP, as demonstrated in some ultrafiltration studies (Cheryan, 1986).

At pH 6.00, the starting permeate flux was 75% higher, when UTMP was doubled (Figure 5). There was much smaller increase in permeate flux at CF = 2 and 4, when UTMP was increased. Due to higher slope of these permeate flux curves (when plotted against CF); there was a cross-over point around CF 6 ($P > 0.05$). Around this CF, the permeate flux rates at constant CFVs were very similar to each other. However, at high CF, the effect of increasing UTMP became negative. For example, at pH 6.50, at CF of 8x, permeate flux was 42% and 27% lower when UTMP was doubled at 5.3 $ms^{-1}$ and 6.3 $ms^{-1}$ respectively. Likewise, at pH 6.00 and CF = 10, the permeate flux was 82% lower at at 5.3 $ms^{-1}$ and 59% at 6.3 $ms^{-1}$, when UTMP was doubled. Except between CF of 6 to 7, the effect of UTMP on permeate flux at constant CFV was always significant ($P < 0.05$). Tables 7 and 10 show that time taken to reach 8x (CF) could be reduced by up to 18% at pH 6.50 and 27% to reach CF = 10x at pH 6.00 by doubling the UTMP. The data in Tables 6 and 9 (in *Appendix*) also suggests that abysmally low permeate flux at higher UTMPs would require a shut down of process and would prevent any further concentration, whereas in case of lower UTMPs, permeate flux declined more gradually, and was significant enough to allow achieving even higher concentrations. Multiple cleaning cycles were needed to restore clean water flux when high UTMP and low CFV was used, suggesting that membrane was more severely fouled. These results suggest that lower UTMP should be beneficial if

the objective is to achieve very high concentrations in a continuous CFM process at high CFV. In a batch process, highest UTMP required shortest CFM time (Tables 7 and 10 in *Appendix*). If the objective is to only achieve lower CF such as 4 to 5x, it might be economical to use higher UTMPs for continuous CFM process. The effect on quality and suitability of such retentate produced for downstream processing to manufacture cheese is discussed in further detail later in this chapter. Guiziou et al. (2004) also recommended higher TMP to achieve higher permeate flux for CF of up to 2 during reverse osmosis. Nakanishi and Kessler (1985) found that reducing TMP during UF improved the rate of removal of deposited layer during cleaning considerably. Rapid decline in permeate flux was also reported by Guiziou et al. (1999) and Marshall et al (1997), at higher TMPs, who also suggest that it is important to optimize the permeate flux and TMP, below which the driving force is too slow and above which, the increase in fouling causes a large reduction in permeate flux.

### 2.4.3. Effect of pH on permeate flux

It has been reported that in situ reduction of pH during CFM of skim milk invariably strips bound calcium from casein, which results in membrane fouling and a lower permeate flux (Marshall et al. 2003). Since proteins are believed to be globular in their native state, as pH is lowered towards its iso-electric point, the molecule starts to unfold, and due to change in ionic strength, bound calcium is stripped away (Walstra and Jenness, 1984). It was observed in our experiments that under similar operating conditions, casein concentration in gel layer was up to 35% higher at lower pH (Figures 6 and 7). Factors such as pH, ionic strength and electric charge of feed material were significant factors to permeate flux, because of their effect on charge on membrane, charge on particles, and conformation and stability and adhesiveness of molecules (Vyas et al. 2000). This is evident when comparing Figures 4 and 5. Only

around 7 and 8x, the permeate flux becomes similar, and this is due to the fact that experiments at pH 6.00 were run on a higher surface area membrane (more available fouling area per kg of feed), while the amount of starting raw material was the same. Therefore, the area available for fouling per kg of feed was greater in latter case, and it can be concluded that similar trend of lower permeate flux at lower pH could be expected through out the experiment, if the membrane area were similar. Kulozik (1998) reported that removal of calcium, both soluble and bound by ion exchange, resulted in disintegration of casein micelle structure, causing a very dense deposited protein layer consisting of casein sub-micelles and significant reduction in permeation rate.

### 2.4.4. Effect of CFV and TMP on Chemical Composition at pH 6.50

In this study it was observed that changing CFV and TMP had a significant effect on the amount of solids and type of solids retained in highly concentrated retentates (CF = 8) during CFM. A complete chemical analysis was done on final retentate samples collected at 8x and the results are shown in Table 2. The range of total solids was from 25.93% to 26.68%. Increasing CFV significantly ($P < 0.05$) reduced total solids in retentate and increasing UTMP significantly increased ($P < 0.05$) the total solids in retentate. The results suggest that lower fouling due to high shear at even 20% higher CFVs allowed more permeation of soluble solids such as minerals (ash) and serum proteins, while increasing UTMP caused severe fouling and negatively impacted protein permeation. Fat was completely retained in all experiments due to its size. An increase in CFV significantly caused lower total protein and true protein in the retentate. Also, increase in UTMP significantly ($P < 0.05$) increased total protein and true protein in the retentate, which contradicts Samuelsson et al. (1997), who reported that retention of WP was independent of TMP in the range of 10-190 kPa at CF of

1.15. Guiziou et al. (2004) reported that increasing TMP was likely responsible for the compression of membrane deposits and lower transmission of alpha lacta albumin and beta lacta globulin and lower overall WP transmission during crossflow microfiltration of reconstituted skim milk CF up to 2. This reduction of true protein in our studies was not due to casein as the difference across various treatments was insignificant ($P>0.05$). The change in true protein levels was due to change in WP. Increase in CFV caused up to 44% lower WP level in final retentate. Higher CFV improved mass transfer and reduced the formation of thick gel layer at membrane surface, that would restrict the permeation of WP. Also, increase in UTMP at each constant CFV led to increased WP level by up to 25%. Samuelsson et al (1997) reported lower total protein and WP in retentate when CFV was increased, while casein remained unchanged. This would also explain why increasing UTMP caused severe fouling and low permeate flux towards the end of the experiment, because WP contributed to membrane resistance. Variation in total calcium levels across different treatments was also insignificant ($P>0.05$). This was not surprising, as most calcium is believed to be bound with casein, and since the casein was completely retained in the retentate, corresponding levels of calcium were also similar.

### 2.4.5. Effect of CFV and TMP on Chemical Composition at pH 6.00

Overall the effect of CFV and UTMP on each component was similar at both pH levels except % total calcium. As was the case at higher pH, changing CFV and TMP had a significant effect on the amount of solids and type of solids retained in highly concentrated retentates (CF = 10) during CFM. A complete chemical analysis done on final retentate samples is shown in Table 3. The range of total solids obtained in final retentate was from 27.63% to 28.61%. Increasing CFV significantly ($P < 0.05$) reduced total solids in retentate and increasing UTMP significantly increased ($P <$

**Table 2. Effect of CFV and UTMP on final Retentate Composition (CF = 8) during CFM of Skim milk at pH = 6.50**

| % Component | Skim Milk | Operating Conditions (CFV x UTMP) for CF = 8 Retentate | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | 5.3 $ms^{-1}$ 68.9kPa | 5.3 $ms^{-1}$ 103.4kPa | 5.3 $ms^{-1}$ 137.9kPa | 5.8 $ms^{-1}$ 68.9kPa | 5.8 $ms^{-1}$ 103.4kPa | 5.8 $ms^{-1}$ 137.9kPa | 6.3 $ms^{-1}$ 68.9kPa | 6.3 $ms^{-1}$ 103.4kPa | 6.3 $ms^{-1}$ 137.9kPa |
| Total Solids [a b c] | 8.85 | 25.93 | 26.29 | 26.68 | 25.79 | 26.02 | 26.41 | 25.63 | 25.84 | 26.12 |
| Ash [a b c] | 0.66 | 2.13 | 2.22 | 2.35 | 2.07 | 2.14 | 2.26 | 2.04 | 2.10 | 2.17 |
| Fat [a] | 0.05 | 0.43 | 0.43 | 0.42 | 0.43 | 0.43 | 0.43 | 0.43 | 0.43 | 0.43 |
| Total Protein [a b c 1] | 3.21 | 18.11 | 18.42 | 18.64 | 17.86 | 18.11 | 18.46 | 17.61 | 17.87 | 18.23 |
| Total Protein [a b c d 1] | 36.27 | 69.84 | 70.06 | 69.87 | 69.25 | 69.60 | 69.90 | 68.71 | 69.16 | 69.79 |
| Casein [a] | 2.43 | 16.21 | 16.18 | 16.16 | 16.20 | 16.20 | 16.18 | 16.19 | 16.19 | 16.18 |
| Whey Protein [a b c 2] | 0.66 | 1.83 | 2.15 | 2.40 | 1.55 | 1.79 | 2.13 | 1.36 | 1.58 | 1.96 |
| Whey Protein [a b c d 2] | 7.46 | 7.06 | 8.18 | 9.00 | 6.01 | 6.88 | 8.07 | 5.31 | 6.11 | 7.50 |
| True Protein [a b c] | 3.09 | 18.04 | 18.33 | 18.56 | 17.75 | 17.99 | 18.31 | 17.55 | 17.77 | 18.14 |
| True Protein [a b c d] | 34.92 | 69.57 | 69.72 | 69.57 | 68.83 | 69.14 | 69.33 | 68.47 | 68.77 | 69.45 |
| Calcium [a] | 0.12 | 0.793 | 0.791 | 0.793 | 0.788 | 0.789 | 0.794 | 0.787 | 0.787 | 0.787 |
| Lactose [a] | 4.93 | 5.26 | 5.22 | 5.27 | 5.43 | 5.34 | 5.26 | 5.55 | 5.44 | 5.29 |

[a] n = 2, $P < 0.05$

[b] Significant difference in Retentate Component when UTMP is changed at constant CFV, $P < 0.05$

[c] Significant difference in Retentate Component when CFV is changed at constant UTMP, $P < 0.05$

[d] Dry Basis

[1] Total Protein = Total Nitrogen x 6.38

[2] Whey Protein = (Non Casein Nitrogen – NPN) x 6.38

**Table 3. Effect of CFV and UTMP on final Retentate Composition (CF =10) during CFM of Skim milk at pH = 6.00**

| % Component | Skim Milk | Operating Conditions (CFV x UTMP) for CF = 10 Retentate | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|
| | | 5.3 $ms^{-1}$ 68.9kPa | 5.3 $ms^{-1}$ 103.4kPa | 5.3 $ms^{-1}$ 137.9kPa | 5.8 $ms^{-1}$ 68.9kPa | 5.8 $ms^{-1}$ 103.4kPa | 5.8 $ms^{-1}$ 137.9kPa | 6.3 $ms^{-1}$ 68.9kPa | 6.3 $ms^{-1}$ 103.4kPa | 6.3 $ms^{-1}$ 137.9kPa |
| Total Solids [a b c] | 8.85 | 27.86 | 28.23 | 28.61 | 27.68 | 27.96 | 28.33 | 27.63 | 27.81 | 28.17 |
| Ash [a b c] | 0.66 | 2.24 | 2.31 | 2.38 | 2.16 | 2.23 | 2.31 | 2.11 | 2.19 | 2.25 |
| Fat [a] | 0.05 | 0.39 | 0.4 | 0.39 | 0.41 | 0.39 | 0.40 | 0.39 | 0.39 | 0.39 |
| Total Protein [a b c 1] | 3.21 | 21.23 | 21.45 | 21.71 | 21.06 | 21.26 | 21.53 | 20.78 | 21.04 | 21.36 |
| Total Protein [a b c d 1] | 36.27 | 76.20 | 75.98 | 75.88 | 76.08 | 76.04 | 76.00 | 75.21 | 75.66 | 75.83 |
| Casein [a] | 2.43 | 19.16 | 19.14 | 19.15 | 19.13 | 19.15 | 19.14 | 19.15 | 19.15 | 19.16 |
| Whey Protein [a b c 2] | 0.66 | 1.98 | 2.22 | 2.47 | 1.84 | 2.04 | 2.29 | 1.56 | 1.78 | 2.08 |
| Whey Protein [a b c d 2] | 7.46 | 7.11 | 7.86 | 8.63 | 6.65 | 7.30 | 8.08 | 5.65 | 6.40 | 7.38 |
| True Protein [a b c] | 3.09 | 21.14 | 21.36 | 21.62 | 20.97 | 21.19 | 21.43 | 20.71 | 20.93 | 21.24 |
| True Protein [a b c d] | 34.92 | 75.88 | 75.66 | 75.57 | 75.76 | 75.79 | 75.64 | 74.95 | 75.26 | 75.40 |
| Calcium [a b] | 0.12 | 0.520 | 0.509 | 0.509 | 0.502 | 0.514 | 0.511 | 0.493 | 0.491 | 0.492 |
| Lactose [a] | 4.93 | 4.00 | 4.07 | 4.13 | 4.05 | 4.08 | 4.09 | 4.35 | 4.19 | 4.17 |

[a] n = 2, $P < 0.05$

[b] Significant difference in Retentate Component when UTMP is changed at constant CFV, $P < 0.05$

[c] Significant difference in Retentate Component when CFV is changed at constant UTMP, $P < 0.05$

[1] Total Protein = Total Nitrogen x 6.38

[2] Whey Protein = (Non Casein Nitrogen – NPN) x 6.38

0.05) the total solids in retentate, and most of this change was due to serum proteins. Fat was completely retained in all experiments due to its size. Am increase in CFV significantly lowered the total protein and true protein levels in the retentate. Higher CFV was effective in improving mass transfer and therefore reduced serum protein retention. Also, increase in UTMP significantly increased both total protein and true protein in the retentate, where increased pressure caused pore plugging which increased serum protein retention on as is basis and also on dry basis. Variation in level of casein across all treatments was insignificant ($P>0.05$). Increase in CFV led up to 27% lower WP levels in retentates. Also, increase in UTMP at constant CFV led to 33% higher WP level in retentates. Variation in total calcium levels across different treatments was significant ($P<0.05$) at low pH. Due to lower pH, more bound calcium was dissolved from casein to ionic form and was permeated through the membrane. Except two outliers ($P<0.05$), the trend indicated that higher CFV at constant UTMP led to more than 5% lower calcium levels in retentates. However, the effect of UTMP on calcium level in final retentates at constant CFV was seen as insignificant ($P>0.05$).

### 2.4.6. Effect of CFV, UTMP and pH on WP to True Proteins ratio

It is noted above that WP retention could be changed significantly (up to 33%) by changing CFV and UTMP. When WP is expressed as a percentage of true proteins, the variation, depending on operating conditions, is also magnified. As shown in Figure 10, higher CFV can lead to lower concentration of WP, as a part of total protein, i.e. higher casein %, and it is true at pH 6.50 (CF = 8x) or pH 6.00 (CF = 10x). Effect of CFV and UTMP on WP-casein ratio was greater at pH 6.50. Increase in CFV lowered the WP-casein ratio between 25 to 18% at pH 6.50 and between 21to 16% at pH 6.00. Increase in UTMP increased WP-casein ratio between 31 to 44% at pH 6.50 and 25 to

33% at pH 6.00. Therefore, CFV and UTMP could be used as an effective tool to control WP to casein balance, which could come in handy during downstream processing such as cheese making. While retention of WP can have economical advantages by enhancing the cheese yields, it could also lead to significant functional defects such as poor melt and poor stretch in mozzarella cheese (Brandsma and Rizvi, 2001).

### 2.4.7. Effect of CFV, UTMP and pH on calcium to True Protein ratio

The ratio of calcium to true protein increased with increase in CFV and decreased with increased in UTMP at pH 6.50 (Figure 11.). Although the differences were not significant, the effect of CFV and UTMP followed a similar trend at lower pH (6.00). Although casein was never fully retained in all experiments, WP retention decreased in both cases with higher CFVs. So when the pH was not lowered, no calcium was solubilized from protein and the net effect was that concentration of milk led to increase in % calcium expressed as a fraction of true protein at CF = 8. It is expected that at higher concentrations, the calcium concentration would further increase as casein would still be retained but WP would permeate partially, lowering the true protein level. By lowering the pH, the level of ionic calcium in retentate increased and hence more calcium was permeated. This led to more than 50% lower calcium to true protein ratio at higher concentration (10x) when similar conditions were compared at pH 6.50 and 6.00. This indicates that more calcium was solubilized form casein at pH 6.00 and was therefore permeated instead of being retained at pH 6.50, where it is bound with casein. This information is useful because it is well known that higher calcium levels in fortified milks would increase rennet action, create firmer curd and increase fat loss due to curd shattering and produce cheese which would under melt. Such cheese defects can be prevented by using retentate made by CMF at low pH and

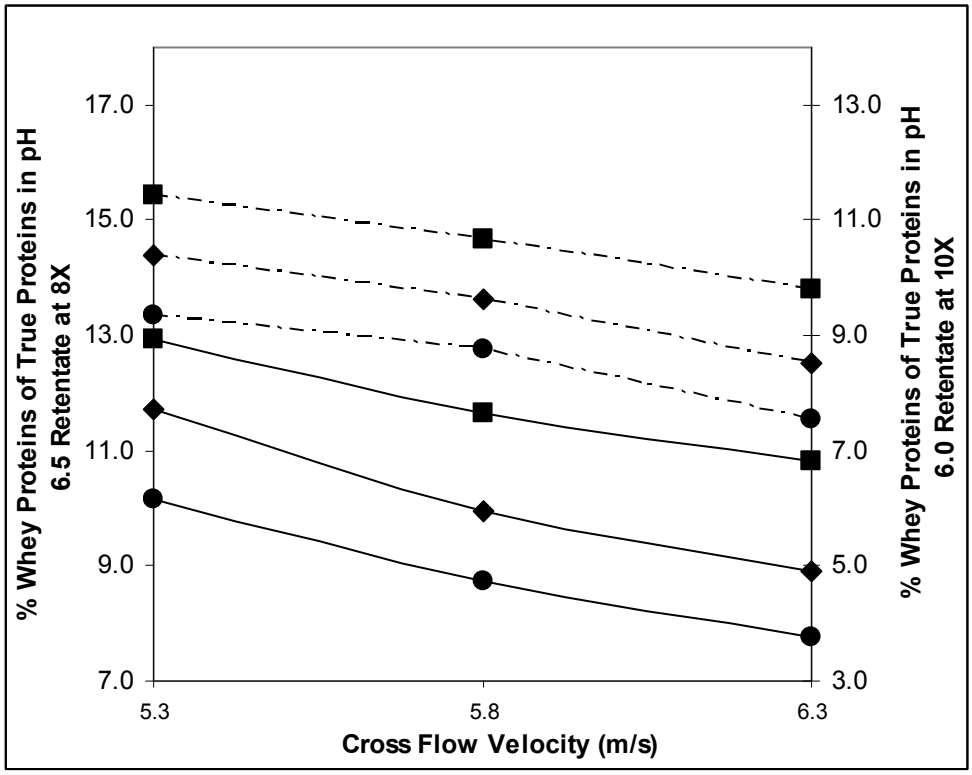


**Figure 10. Effect of CFV x UTMP on % Whey Proteins of True Proteins in Retentate (● = 68.9 k.Pa, ◆ = 103.4 k.Pa, ■ = 137.9 k.Pa, —— = pH 6.50 Retentate, ------ = pH 6.00 Retentate)**

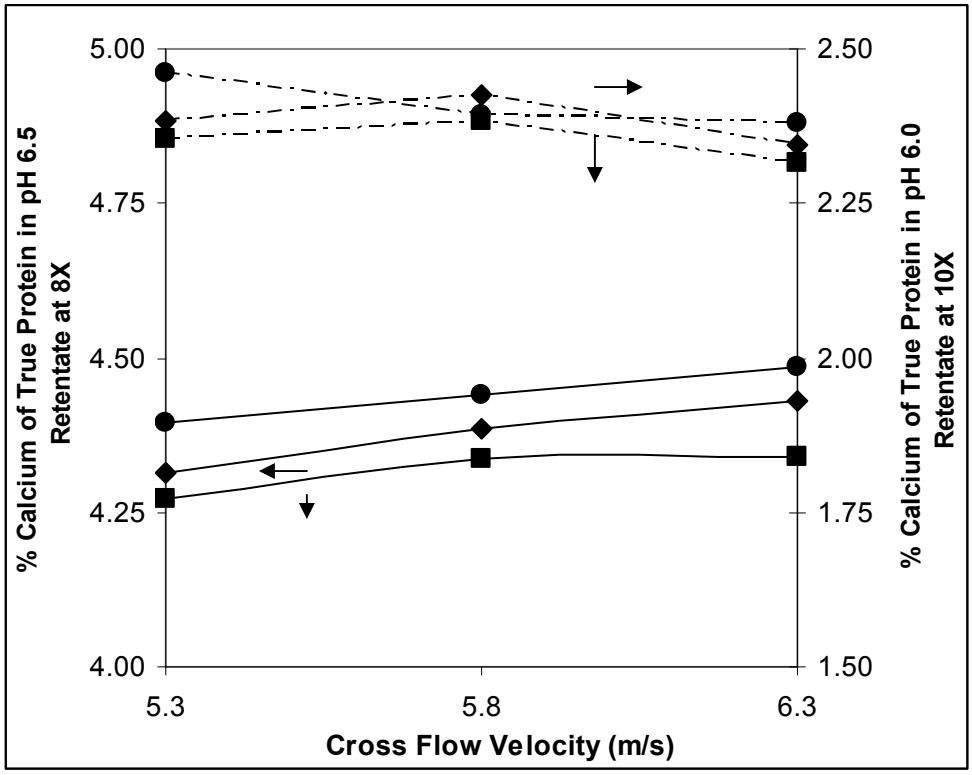


**Figure 11. Effect of CFV x UTMP on % Calcium of True Protein in Retentate (● = 68.9 k.Pa, ◆ = 103.4 k.Pa, ■ = 137.9 k.Pa, —— = pH 6.50 Retentate, ------ = pH 6.00 Retentate)**

very high concentrations, where there is lesser whey syneresis, as explained by Korat and Rizvi (2004).

### 2.4.8. Effect of CFV, UTMP and pH on Calcium to Casein ratio

Change in ratio of total calcium and casein by changing CFV and UTMP was insignificant at higher pH (6.50), because most of the calcium in milk is bound to casein and casein was never fully retained during CMF (Figure 12). Therefore, it can be expected that calcium to casein ratio would remain constant during CFM of skim milk at pH close to normal milk (6.50), which is important for predicting rennet activity during cheese make. However, during CFM at lower pH (6.00), calcium is stripped from casein, and ionic calcium is free to permeate. It was seen that increase in CFV could significantly improve permeate flux at higher concentrations, therefore allow more fractionation and hence more calcium removal. There was no definite trend in the effect of UTMP on total calcium to casein ratio. It can therefore be concluded that CFM at higher CFV and at low pH (6.0) will produce concentrated retentates with lower calcium to casein ratio than standard non-acidified retentates, which are more suitable for cheese make as described before.

## 2.5. CONCLUSIONS

Consistently higher permeate flux was obtained at higher CFV suggesting that higher shear rates at membrane surface results in reduction of gel layer concentration and aided in back transport of fouling layer into the feed stream. Increasing CFV by approximately 20% (from 5.3 to 6.3 $ms^{-1}$) at startup increased starting permeate flux by up to 34% at constant UTMP. As CFV was increased, concentration polarization was reduced at the membrane surface due to better mass transfer, and the molecules rejected by the membrane were more rapidly transported back to the retentate stream.

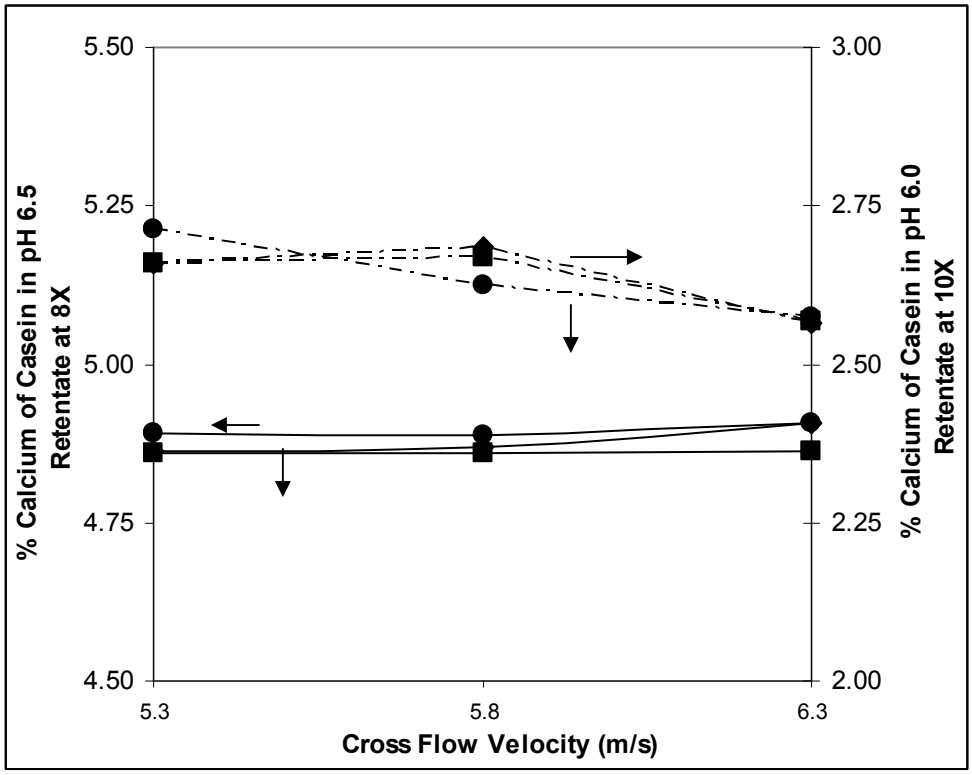


**Figure 12. Effect of CFV x UTMP on % Calcium of Casein in Retentate (● = 68.9 k.Pa, ◆ = 103.4 k.Pa, ■ = 137.9 k.Pa, —— = pH 6.50 Retentate, ------ = pH 6.00 Retentate)**

By doubling UTMP (from 68.9 to 137.9 kPa), starting permeate flux increased by an average of 20% at pH 6.50 and 60% at pH 6.00. This difference in flux at different pH was more dramatic probably due to change in protein-protein, protein-mineral interactions within the retentate and between retentate and fouled membrane because at lower pH, more calcium was dissolved from casein. As UTMP was increased, permeate flux increased initially but fell below a critical level at higher concentrations (7 to 8x). There was up to 97% decline in permeate flux from start to end, due to CF, with typically 60% decline coming after 6x. It can be concluded that higher CFV in combination with higher UTMP is desirable to achieve CF up to 4 and a combination of high CFV with low UTMP would make it possible to achieve higher CF such as 10x for continuous operations, as evident by shorter cleaning cycles at lower UTMP and higher CFV. Designer retentates can be produced with required component balance (casein: whey protein: calcium: lactose), suitable for any downstream processing by manipulating the main operating conditions, i.e. CFV, UTMP and pH at the same time producing excellent quality permeate stream which is mostly sterile. The information can be significant where macro components of milk such as WP and calcium play a significant role in final product functionality such as different types of cheese, RTD (Ready To Drink) beverages, high protein bars. High CFV is always more desirable at each UTMP since higher flux was always obtained by using high CFV, which results in high shear and prevents cake build up on the membrane surface and aids in WP and calcium permeation. Higher UTMP (137.9 vs. 68.9 kPa) will help to finish CFM batch run in shortest time and highest flux up to 6-7x, but will result in severe fouling and a sharp drop in flux if the process is continued to higher concentrations (8-10x) and increased WP retention.

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# CHAPTER THREE

# PROCESS ANALYSIS OF CROSS FLOW MICROFILTRATION FOR SELECTIVE CONCENTRATION OF SKIM MILK PROTEINS

## 3.1. ABSTRACT

This study reports the effects of concentration factor, CFV, and UTMP on energy requirements for selectively concentrating pasteurized skim milk up to 8 times using CFM (0.2μm) at pH 6.50 and at 50$^{o}$ C. Volumetric concentration of 8x was achieved at each combination of 3 CFVs and 3 UTMPs; permeate flux and longitudinal pressure drop were recorded at each CF, and power consumption was calculated. A transition from turbulent to laminar flow was observed around 8x, and retentate behavior became shear thinning around 4 to 6x. Increase in CFV from 5.3 to 6.3 ms$^{-1}$ increased permeate flux and reduced over all power consumption by 25%. Increase in UTMP (68.9 to 137.9 kPa) enhanced the starting permeate flux by 20% and lowered the corresponding power consumption by 18%, but beyond 6x, higher UTMP led to lower permeate flux and double the power consumption. Overall energy consumption was always lower due to shorter CFM process when skim milk was microfiltered to 8x at higher CFV and higher UTMP.

## 3.2. INTRODUCTION

Since the 1980s, CFM has emerged as an industrially feasible technology for milk and milk component processing thanks to the development of new ceramic membranes with multi-channel geometry to be used in cross/tangential flow mode. The new membrane modules also have a highly permeable support, which has allowed the hydraulic concept called UTMP to be put into effect, making high CF achievable and

economically feasible (Maubois, 2002). CFM has been investigated for uses in the pharmaceutical industry (electro coagulation for virus removal, aseptic harvesting of bacteria, antibody and enzyme recovery, high cell density cultivation, washing cryopreserved blood products); the chemical industry (water treatment, removal of surfactants, paper pulp processing, bioreactors and fermentors) and the food industry. Novel food industry uses involve fractionation and concentration of dairy liquids into unique value added streams, hydrolysates refining, soft-oil decolorization and purification, clarification & concentration of wine, beer, juices, vinegar and recovering aromatic compounds from liquid food streams (Cheryan, 1998). The time honored and tested applications involving membrane processing of milk have introduced a plethora of refined proteins and virgin dairy protein streams and opened up several commercial uses (Korat and Rizvi, 2004; Maubois 2002). Cheese making by using concentrated milk has been of interest to the food industry for well over two decades. As more and more cheese plants incorporate membrane processing in cheese manufacture to standardize cheese milk and increase total solids (Mistry 2001), the need to analyze this process itself becomes more critical so that membrane processing can become integrated with other dairy manufacturing operations. Microfiltration of skim milk, especially for mozzarella cheese making is more advantageous as compared to ultrafiltration, due to the suitability of the retentate composition obtained (Korat and Rizvi, 2004).

Selective concentration of skim milk using CFM retains almost all casein and partially retains serum proteins. A high degree of selective concentration (8 to 10x) is needed to considerably reduce the equipment size and labor needed for downstream processing and possibly eliminate whey-processing equipment (Korat and Rizvi, 2004). CFM can be a very useful tool to optimize mozzarella cheese manufacture by reducing capital investment, but no study to date has been done to evaluate the CFM process to

concentrate skim milk to very high concentrations. No study using non-UTMP to concentrate skim milk has ever reported achieving CF higher than 4 to 5x. UTMP was pioneered and patented by Sandblom in 1974 and made it possible to reap the benefit of efficient particle back transport from the membrane wall at high axial wall shear rates (high CFVs) while maintaining low TMP in the pressure dependent regime (Cheryan, 1998). The chief advantage of CFM process in UTMP mode is the uniform fouling of the membrane, uniform erosion of solute particles from the membrane surface (Vadi and Rizvi, 2001) and better solute transport through the membrane (Berre and Daufin, 1996) which permits achieving a higher CF. Fouling is mainly characterized by adsorption and pore-plugging (Merin and Daufin, 1990). Their studies have confirmed that there is a critical permeation flux in cross flow microfiltration, under which little or no fouling takes place and above which fouling increases sharply. However, that critical flux is very much lower than the economical flux, and it would need tediously long hours to achieve significantly high CF such as 8 to 10x. Because milk is a complex biological fluid, microfiltration is accompanied by a continuous increase in the fouling layer, and the operating conditions largely determine the extent of fouling (Vyas et al., 2000b; Berre and Daufin, 1996). With continuous increase in fouling, the resistance offered by the membrane increases and hinders microfiltration at higher CF. Therefore more shearing action is often required to minimize the formation of cake layer on the membrane surface (Berre and Daufin, 1996). The major energy cost for cross-flow UTMP microfiltration is pumping energy spent on recirculating feed and permeate, the cooling water used to maintain the temperature of recirculating retentate, and the cleaning solutions used later to recover the permeate flux lost due to fouling. The process is expected to become energy intensive at higher concentrations (6x and above) because of the sharp increase in viscosity due to increase in total solids and continuous fouling of the membrane.

All process applications of membrane filtration serve an economic objective, and therefore the product output should be optimized under different objective functions such as concentration levels, maximum flux, minimum energy cost and maximum byproduct benefits. Major factors that affect the performance of a microfiltration system are CFV, TMP, CF and temperature and feed composition. In this study we report the effects of CF, cross flow velocity (CFV), and UTMP on energy requirements for selectively concentrating skim milk components (casein) up to 8 times using CFM.

## 3.3. MATERIALS AND METHODS

### 3.3.1. Microfiltration System

The cross flow microfiltration assembly (Figure 13.) used in this study had two 0.2-micron membrane elements (1P/R19-40, US Filter Corp., Warrendale, PA). Each membrane element had 19 channels, with a diameter of 4 mm. The length of each element was 850 mm, yielding a total surface area of 0.4 $m^2$. Both elements were placed in separate parallel housings. The inlet and outlet pressures on both retentate and permeate sides were measured with pressure gauges (Anderson Instrument Co.), and could be controlled by flow control valves. The feed flow rate was kept constant with the help of flow meters (Series 55-200, Wallace & Tiernan, Belleville, NJ) attached at the inlets of both membrane elements. The pump used for retentate circulation was centrifugal and water-cooled (7.5HP, Reliance Electric Co.), and the permeate circulation pump was centrifugal and air-cooled (5 HP, Reliance Electric Co.). A coaxial heat exchanger brought the temperature of the retentate down during its return to feed tank, because retentate heated up gradually due to friction. In addition, a constant head was maintained on permeate side with a small overhead tank (12 kg), and permeate flow rate was adjusted to achieve uniform trans membrane

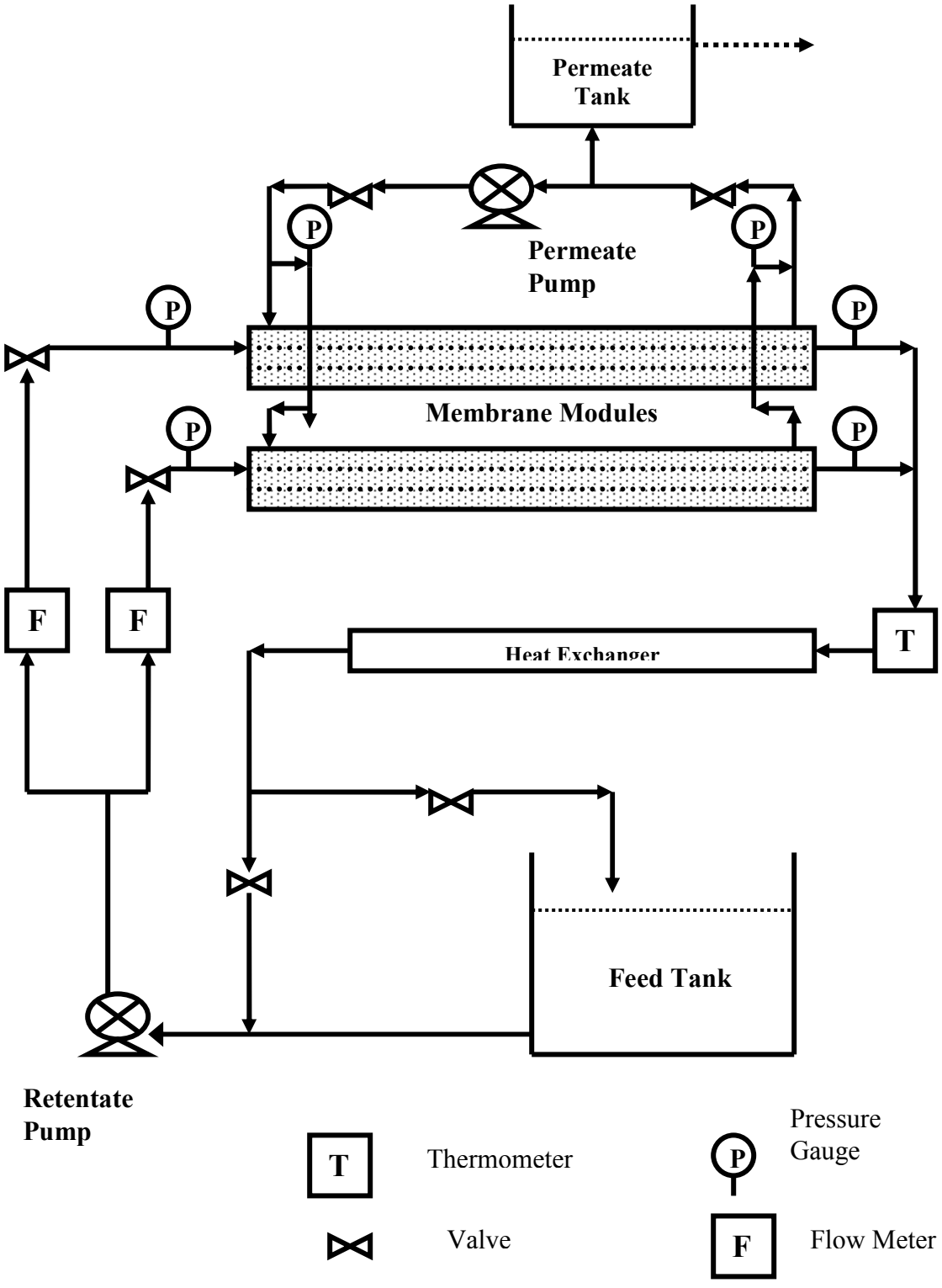


**Figure 13. Schematic of Cross Flow Microfiltration System**

pressure. Any net overflow was measured as the flux of the system. The system was operated in batches, and the dead volume of the system was 16 kg.

### 3.3.2. CFM Process

Pasteurized skim milk (72$^{o}$C for 15 s) was microfiltered using CFM (0.2 μm pore size) at 50$^{o}$C at three CFVs (5.3, 5.8 and 6.3ms$^{-1}$) and three UTMPs (68.9, 103.4 and 137.9kPa) in duplicate runs in a 3x3 factorial design. Volumetric CF of 8x was achieved at each combination of CFV and UTMP at pH 6.50. Pressure drop across the membrane length was compensated by adjusting the permeate flow to achieve uniform trans membrane pressure throughout. For example at CF = 4, the retentate/permeate pressures were 448.2/310.3 kPa at the inlet and 337.9/200.0 kPa at the outlet, giving a UTMP of 137.9 kPa and a longitudinal pressure drop of 110.3 kPa. To lower the pH during CFM, a known amount of glucono-delta-lactone was added in the beginning. Retentate temperature was maintained at 50 ± 1$^{o}$C by recirculating cold water at 4$^{o}$ C, because as the retentate was concentrated, it became more viscous and therefore heated up due to friction losses. The permeate flux (or filtrate rate), expressed in KMH (kg-m$^{-2}$-h$^{-1}$) was measured every 10 min, and the pressures and flow rates were closely monitored. Samples of retentate and permeate were collected and analyzed for chemical composition.

### 3.3.3. Cleaning

After the end of each experiment, washing fouled membranes with cleaning solutions restored clean water flux. After rinsing with regular water, first a 2% w/w NaOH solution at 70 $^{0}$C was circulated for 1.5 hrs without opening the permeate line. Then permeate flow valve was opened to clean any plugged membrane pores, and the solution was circulated for another 1.5 hrs though the membrane pores. In the end, the

hot alkali solution was pushed to the permeate tank, and reverse circulation was done for 45 min. These three cycles were also repeated for 2% $HNO_3$ w/w at 70$^o$ C after rinsing the alkali out. Finally, the membrane was rinsed and the water flux at 137.9 kPa (9000 KMH) was checked before the start of next experiment. During cleaning, the temperature was increased or decreased 1 $^o$ C / min to prevent heat shock to the membranes.

### 3.3.4. Compositional Analysis

Total solids were determined using forced oven drying (AOAC, 1995) and fat by Mojonnier ether extraction (AOAC, 1995). Ash was determined by drying samples in a forced air oven at 100$^o$C and then placing the sample dish in a muffle furnace for 20 h at 550$^o$C. Nitrogen content (total nitrogen (TN), non-casein nitrogen (NCN), non-protein nitrogen (NPN)) in samples was determined by Kjeldahl method (AOAC, 1995). Total protein was calculated by multiplying the total nitrogen by 6.38. True protein was calculated as (TN - NPN) x 6.38 and casein as (TN - NCN) x 6.38. Whey Protein (WP) was calculated as (NCN – NPN) x 6.38. Total calcium was measured by atomic absorption spectroscopy analysis, as described by Metzger et al (2000). Finally, lactose was calculated by difference. All samples were analyzed in duplicates.

### 3.3.5. Density and Flow Property Measurements

The density of retentates was determined in duplicate with a volumetric pycnometer (Weissberger, 1971). Flow properties of skim milk and retentates were determined in duplicate at 50 $^o$C on a Haake RV 100 double concentric cylinder viscometer (Haake Buchler, Saddle Brook, NJ) fitted with NV sensor system. A Haake PG 142 automatic programmer was used to ramp up shear rate ($\gamma$) over the range 0 to 2700 $s^{-1}$ within 4 min. Data was collected through a Yokogawa HR 2400 data logger (Yokogawa

Electric Corp., Tokyo, Japan) and then transferred to Excel 5.0 software (Microsoft Corp., Redmond, WA) for further analysis. Shear stress (τ)-shear rate (γ) data was modeled according to the power law equation

$$\tau = K\gamma^{n}, \quad (1)$$

where n is the flow behavior index (-) and K is the consistency coefficient ($Pa.s^n$). With power law parameters, the effective viscosity (μ) was calculated through the equation.

$$\mu = K\gamma^{n-1}, \quad (2)$$

### 3.3.6. Statistics

MINITAB release 9 (Minitab Inc., State College, PA) and Microsoft Excel (Microsoft Corp, Redmond, WA) were used for statistical analysis of the data. Permeate flux during skim milk CFM was measured every 10 min. The effect of CFV was assessed by using 3 different CFV (5.3, 5.8, 6.3 $ms^{-1}$) at fixed UTMP using ANOVA. Similarly, the effect of UTMP was assessed by using 3 different UTMP (68.9, 103.4, 137.9 kPa) at a given CFV using ANOVA. Significant differences were determined at $P < 0.05$. The whole experiment was formulated into a two level 3x3 factorial design. Experiments were performed in random order to negate any carry over effects. After every run was completed, the permeate flux as a function of CF was plotted. A best – fit (polynomial) regression line was drawn through the data set and the instantaneous flux values at each CF were calculated from polynomial equation obtained from the graph. This exercise was repeated for all the runs to get flux values at each CF.

### 3.3.7. Process Modeling

Based on the flow behavior of the retentate the energy consumption calculations were done using two models.

At concentrations less than 8x, the flow behavior resembled a Newtonian fluid. The first model used to calculate the longitudinal pressure drop for a **Newtonian turbulent flow** (CF < 8x) was (Steffe, 1992):

$$\Delta P = \frac{f\rho L Q_{ave.}^2}{m^2 \pi^2 R^5} \tag{3}$$

where ΔP is longitudinal pressure drop across the length (L) of the membrane, ρ is the density, $Q_{ave}$ is average volumetric flow rate, m is number of membrane elements and R is the radius of each element and, $f$ is the friction factor calculated from Equation (8) below.

A second model was used for calculating longitudinal pressure drop for **pseudo plastic laminar flow** (CF of 8x), which is (Steffe, 1992):

$$\Delta P = \frac{2KL}{R}\left(\frac{3n+1}{4n}\right)^n \left[\frac{4Q_{ave}}{\pi R^3}\right]^n \tag{4}$$

where, K is the consistency index, n is the flow behavior index.

Therefore, power consumed in the CFM process was calculated as

$$P = Q_{ave}\Delta P \tag{5}$$

Where, P is the power consumed.

Therefore power consumed in the CFM process for concentrations up to 6x, when the flow behavior was turbulent was calculated from the following derived formula:

$$P = \frac{f\rho L Q_{ave.}^3}{m^2 \pi^2 R^5} \tag{6}$$

The power consumed (P) in the CFM process for concentration of 8x, when the flow behavior was laminar was calculated from

$$P = Q\frac{2KL}{R}\left(\frac{3n+1}{4n}\right)^n\left[\frac{4Q_{ave}}{\pi R^3}\right]^n \tag{7}$$

The friction factor (***f***) was calculated from a general equation for smooth tubes as follows (Steffe, 1992):

$$\frac{1}{(f)^{1/2}} = \frac{1.74}{n^{0.75}}\left[\ln \mathrm{Re}(f)^{1-0.5n}\right] - \frac{0.4}{n^{1.2}} \tag{8}$$

where *Re* is the *Reynolds number* calculated from the commonly used equation for CF < 4x:

$$\mathrm{Re} = \frac{\rho D v}{\mu} \tag{9}$$

For CF >4x, the equation based on power law parameters was used to calculate *Reynolds number* (McCabe et al., 2001):

$$\mathrm{Re} = 2^{3-n}\left[\frac{n}{3n+1}\right]^n \frac{\rho D^n v^{2-n}}{K} \tag{10}$$

where, D is the membrane channel diameter and v is CFV.

All the power consumed as shown in data that follows below is instantaneous and calculated. These power values represent only the retentate side. Power consumed on the permeate side was assumed to be constant due to no apparent change in permeate rheological properties. The power needed to recompress the retentate back into the membrane unit was not included in these calculations.

### 3.4. RESULTS & DISCUSSION

Calculated longitudinal pressure drop values were compared with observed pressure drop in a few cases, and the results are shown in Figure 14. Both values compare very well from skim milk to CF of 4x. After that, the observed longitudinal pressure drop was about 20% higher than predicted. The fact that the pressure values correlate very well in the beginning but not in the end is surprising. One possible reason could be that initially, the deposits on the membrane are extremely thin. It could be assumed that as the concentration increases, the thickness of fouling layer increases. Therefore, a deposit of even a few microns (on a 4mm diameter) in the beginning would not be a significant reduction to cause noticeable difference in calculated and observed pressure drop. As the effective diameter of membranes continues to decrease, the longitudinal pressure drop increases. If the deposits are thicker (50 to 100 microns), a large difference in calculated and observed pressure drop can be expected. Based on our calculations, it is expected that at 8x, the effective diameter was only 3.78mm (instead of 4mm used for calculations). It was assumed using Reynolds number calculations that the flow turned from turbulent to laminar somewhere around after CF = 8 (Table 4). The variance in observed and predicted pressure drop values (Figure 14) indicates that either the flow did not become completely laminar at CF = 8, or the calculated viscosity used in process modeling was much different than the actual viscosity inside the membranes. The effective shear rate in the membranes was at least several times higher (10,600 $s^{-1}$) than the highest shear rate (2700 $s^{-1}$) used in the viscometer measurements. We used the pressure drop equation for Newtonian fluids assuming that our fluid was Newtonian for CF of 6x and lower. This assumption provided us a pressure drop value that was closer to what we observed during experiments (128 to 138 kPa). When an equation for non-Newtonian fluid was used (based on Table 4), the calculated pressure drop was almost 50% lower (66 kPa).

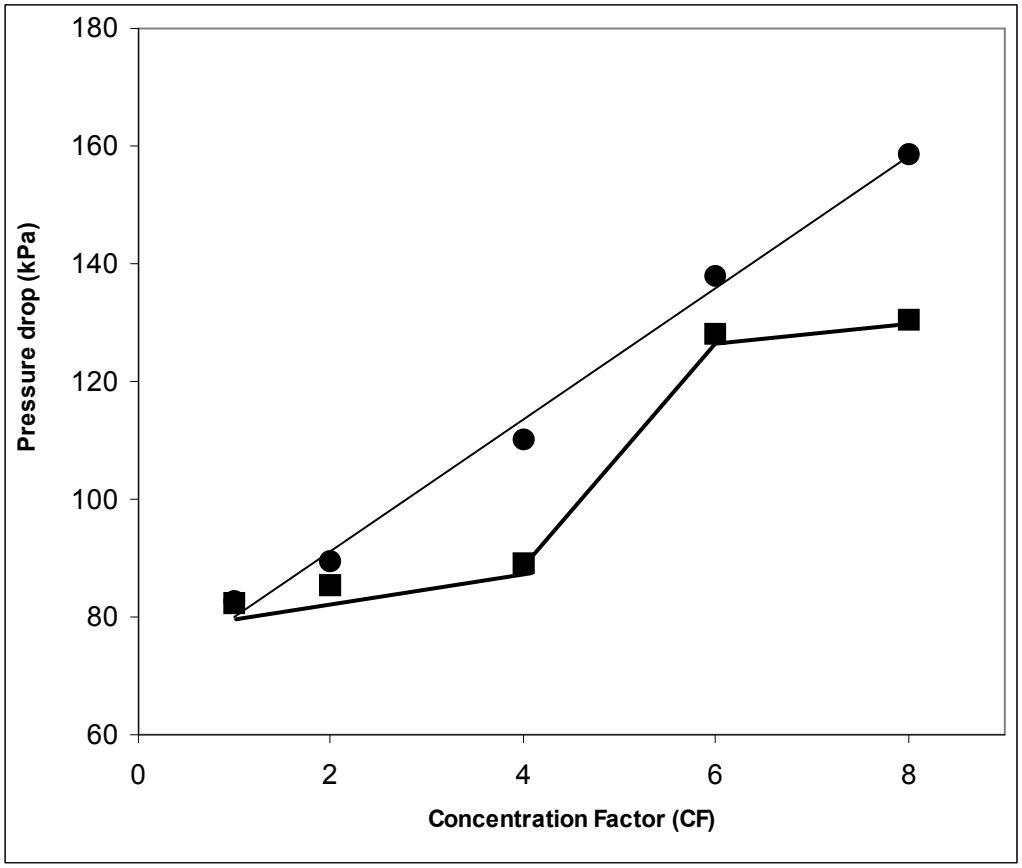


**Figure 14. Observed (●) and Calculated (■) longitudinal pressure drop during cross flow microfiltration of skim milk at 5.3 $ms^{-1}$ and 137.9 kPa**

**Table 4. Physico-chemical properties of skim milk retentate during CFM at 5.3 $ms^{-1}$ and 137.9 kPa**

| CF | Total Solids[1a] % | Reynolds (Re) number [1 b] | Viscosity (cP) [b] | Density (ρ) [1 a] $kg/m^3$ | n-value [1b] | K-value [1b] | Friction factor (f) [b] |
|---|---|---|---|---|---|---|---|
| Skim milk | 8.85 | 16,634 | 1.30 | 1020 | 1.00 | 0.0013 | 0.00676 |
| 2x retentate | 11.51 | 14,826 | 1.47 | 1028 | 1.00 | 0.0020 | 0.00696 |
| 4x retentate | 17.27 | 12,931 | 2.07 | 1040 | 0.89 | 0.0046 | 0.00718 |
| 6x retentate | 23.15 | 3,039 | 9.98 | 1059 | 0.84 | 0.0313 | 0.01014 |
| 8x retentate | 26.68 | 1,570 | 25.7 | 1076 | 0.71 | 0.1993 | 0.01261 |

[1] Mean (n = 2)
[a] Measured
[b] Calculated using Haake Viscometer shear rate ($\gamma = 2{,}700s^{-1}$)

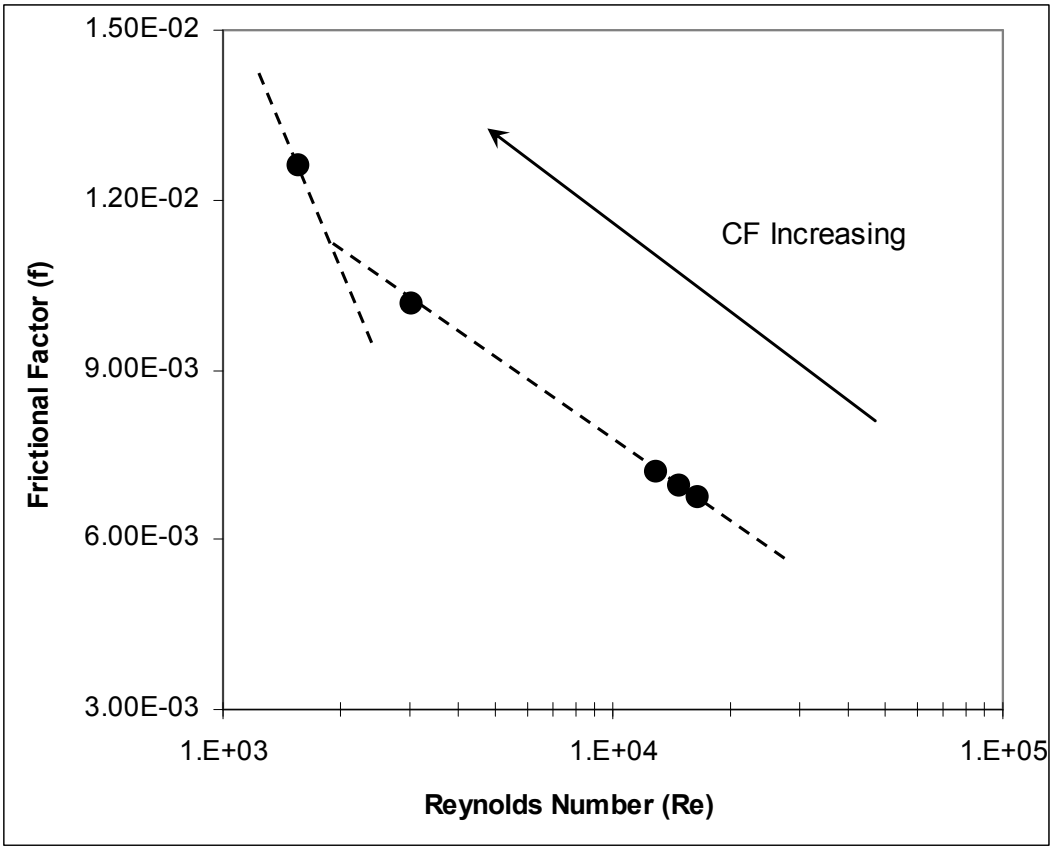


**Figure 15. Variation of Reynolds number and calculated friction factor (*f*) at CF (1, 2, 4, 6, 8)**

An increase in concentration of retentate would increase the viscosity ($\mu$) and the friction factor ($f$) would increase because of lower Reynolds number (Equation 8). Calculated values of frictional factor are listed in Table 4 and are plotted against *Re* in Figure 15. As expected, friction factor increases with CF due to increase in viscosity, suggesting that membrane is fouled and perhaps it's surface becomes rougher during the experiment. The plot (Figure 15) resembles other experimental values of a Newtonian fluid following turbulent flow in a smooth pipe (McCabe et al., 2001). At high CF, effective CFV would be somewhat greater due to reduction in membrane diameter, which contributed to calculated friction factor ($f$) value higher than for a smooth pipe.

In our experiments, we were successful in achieving highly concentrated retentate with approximately 26.68 % total solids at CF of 8x. A detailed comparison of chemical composition of skim milk and final retentate is shown in Table 5. Under ideal conditions, if casein was fully retained, the casein concentration in 8x retentate would be 19.44%, compared to 16.16% as tested. It is possible that some casein is deposited in the membrane pores, and gets lost during the cleaning cycle. Similarly, not all whey protein was transmitted in the permeate. Whey protein concentration at 8x (2.40% as tested), indicates that only 45% whey protein was actually retained.

As indicated by the experiments (Figure 16), the increase in CF is accompanied by a slow decrease (< 10%) in permeate flux up to 4x and faster decline (> 60%) between CF 6x and 8x. This rapid drop in permeate flux indicates concentration polarization and perhaps severe fouling. This could also be partially attributed to the change from turbulent to laminar flow around 8x, because a turbulent flow produces better mass transfer (and higher turbulent Reynolds stress) than laminar flow under the same flow conditions. The drop in flux continues throughout the experiment. Any increase in UTMP caused increase in permeate flux in the beginning. However, if higher UTMP

**Table 5. Chemical Composition of skim milk retentate during CFM at 5.3 $ms^{-1}$ and 137.9 kPa**

| % Component | Skim Milk | 8x Retentate |
|---|---|---|
| Total Solids [a] | 8.85 | 26.68 |
| Ash [a] | 0.66 | 2.35 |
| Fat [a] | 0.05 | 0.42 |
| Total Protein [a 1] | 3.21 | 18.64 |
| Casein [a] | 2.43 | 16.16 |
| Whey Protein [a 2] | 0.66 | 2.40 |
| True Protein [a] | 3.09 | 18.56 |
| Calcium [a] | 0.12 | 0.793 |
| Lactose [a] | 4.93 | 5.27 |

[a] $n = 2$, $P < 0.05$

[1] Total Protein = Total Nitrogen x 6.38

[2] Whey Protein = (Non Casein Nitrogen – NPN) x 6.38

was employed through the entire experiment, it caused a steep decline in permeate flux (at CF > 6x) probably caused by excessive fouling due to pore-plugging and cake layer formation (Merin and Daufin, 1990). The result is even lower permeate flux at higher CF. Use of higher flow velocity resulted in higher erosive action on the fouling layer, better mass transport away from the membrane surface, 33% lower gel layer concentration and higher permeate flux (Vadi and Rizvi, 2001). Although we tried to perform a mass balance for the solids in retentate and permeate, it was difficult to estimate the exact thickness of fouling layer since we did not open the membrane and recover all the retentate.

### 3.4.1. Effect of CF on energy consumption

The energy consumption per unit volume of permeate removed was estimated from the ratio of calculated power consumption and observed permeate flux rate. The permeate flux drops and simultaneously power consumption increases with the CF. The Power/Filtration rate ratio (P/F), (kJ/kg), that gives the energy (kJ) consumed in separating every *kg* of permeate also increases with CF (Figure 16) as expected. Concentrating the skim milk into 2x retentate (30% solids increase), which is a commonly used concentration in the cheese industry for standardization of vat liquid increased the P/F ratio by only 13%. The instantaneous kJ/kg ratio increases several times at higher CF (increasing from 24 kJ/kg for skim milk to 297 kJ/kg for 8x at 5.3 $ms^{-1}$ and 137.9 kPa). This 1100% total increase in power consumption corresponds to roughly three fold increase in total solids of skim milk at CF of 8x and amounts to about $ 0.26 / 1000 kg of permeate removed for skim milk and $ 3.92 / 1000 kg of permeate removed at 8x assuming $ 0.04 / KWH for pumping energy. Such a cost could be well absorbed if the retentate produced was easier to process and permitted better cheese quality, as proven by Brandsma and Rizvi (1999). Power consumed

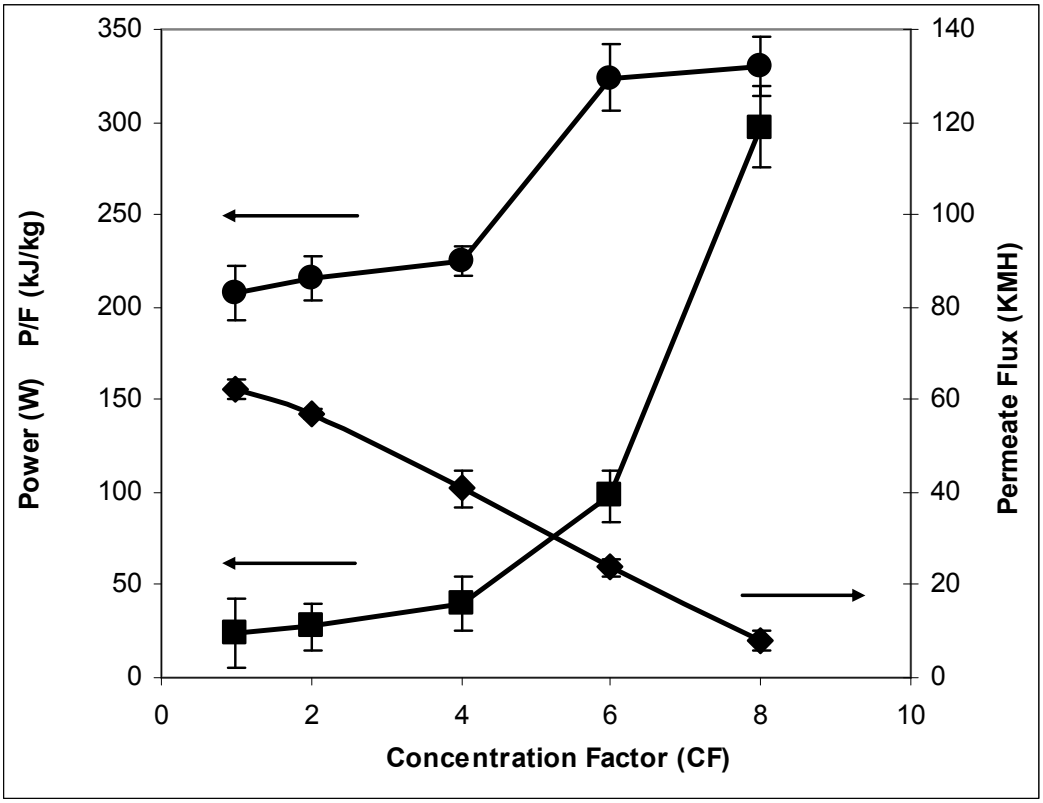


**Figure 16. Effect of CF on power consumed (●), Permeate Flux (◆) and P/F ratio (■) during CFM of skim milk at 5.3 $ms^{-1}$ and 137.9 kPa**

between CF 6 and 8 is relatively very high, therefore more work is done to increase concentration of retentate due to change in its rheological properties. Some energy was also lost continuously when retentate was recycled back to the balance tank at the atmospheric pressure. It is possible that pressurizing the whole CFM system may lead to better economics and could be part of a future study.

### 3.4.2 Effect of CFV on energy consumption

Higher flow rates help reduce the formation of fouling layer on the membrane and help in shear thinning due to reduction in viscosity, since the retentate behaves as power law fluid at concentrations beyond 4x. Higher flow rate also improves mass transfer and leads to better back transport of retained particles into the main retentate stream, reducing gel layer concentration and leading to lower concentration polarization and better permeate flux. It is shown in Figure 5 that higher CFV consumes more power while increasing permeate flux. During microfiltration of skim milk, an increase of flow rate from 5.3 to 6.3 $ms^{-1}$ at 137.9kPa increased energy consumption from 24 to 29.5 kJ/kg for skim milk (Figure 17). A similar increase in flow rate at 6x (CF) resulted in 25% increase in permeate flux from 24 to 30 KMH. This also resulted in slight increase in energy consumption from 98 to 105 kJ/kg (Figure 18). However, at 8x (CF), a similar increase in flow rate led to almost 75% increase in permeate flux (from 8 to 14 KMH) and a significant reduction (25%) in energy consumption ($ 1 per 1000 kg of permeate removed) (Figure 19). This indicated that CFM process at high concentrations is less energy intensive at higher cross flow rates. It is obvious that there would be a limit to the increase in flow rate that can be achieved in any set up. The optimum flow rate should be determined in any given microfiltration equipment to get a reasonable filtration rate without excessive power consumption, and the recommended flow rate would be the highest CFV that

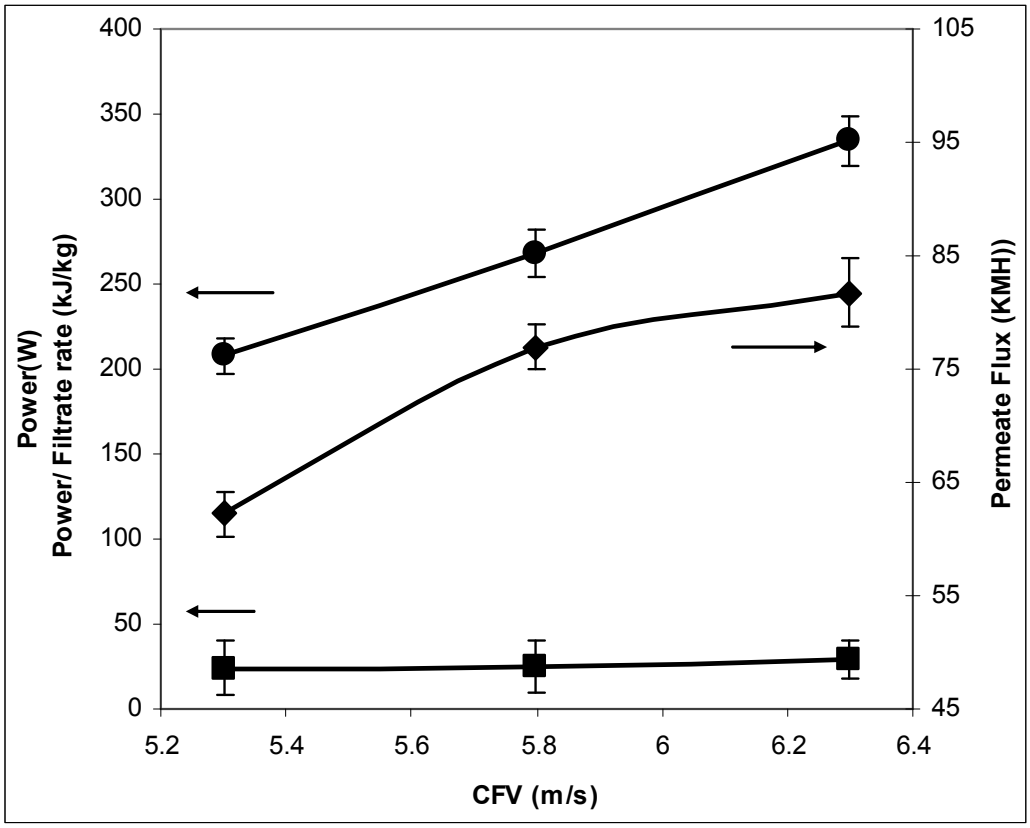


**Figure 17. Effect of CFV on power consumed (●), Permeate Flux (◆) and P/F ratio (■) during CFM of skim milk at 137.9 kPa**

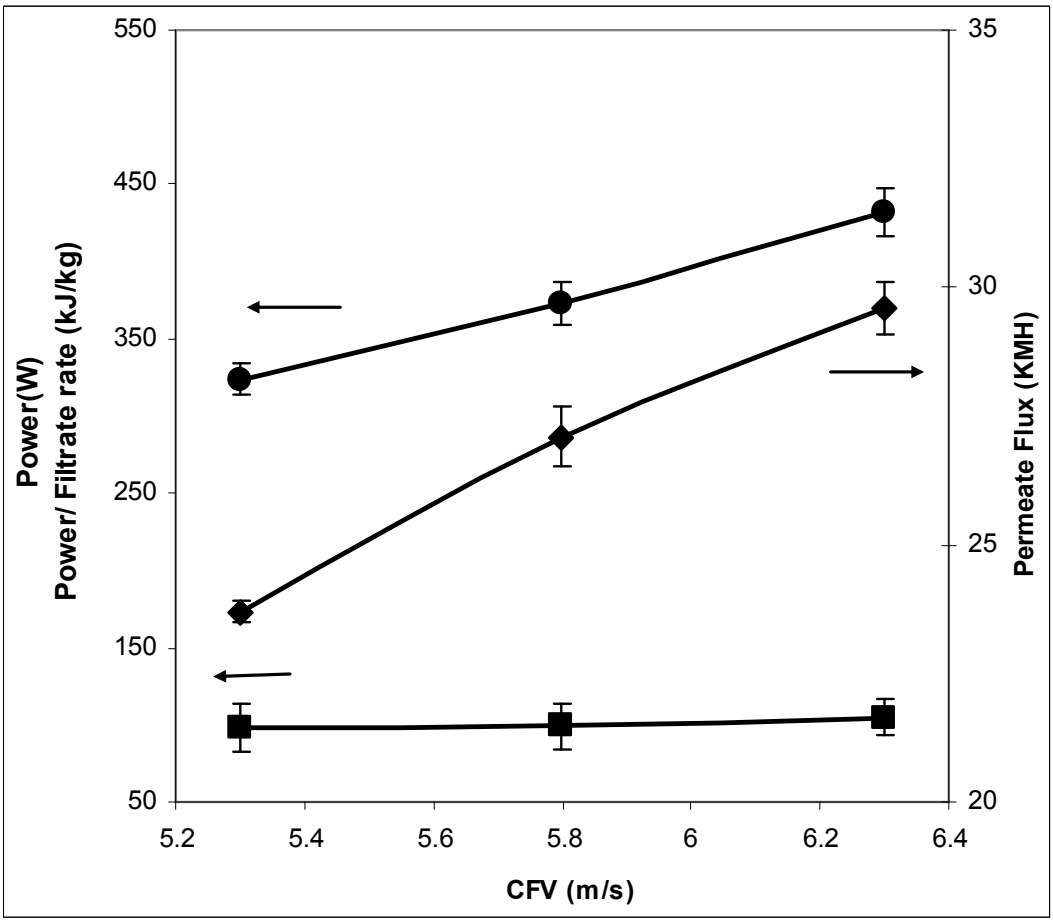


**Figure 18. Effect of CFV on power consumed (●), Permeate Flux (◆) and P/F ratio (■) during CFM of 6x retentate at 137.9 kPa**

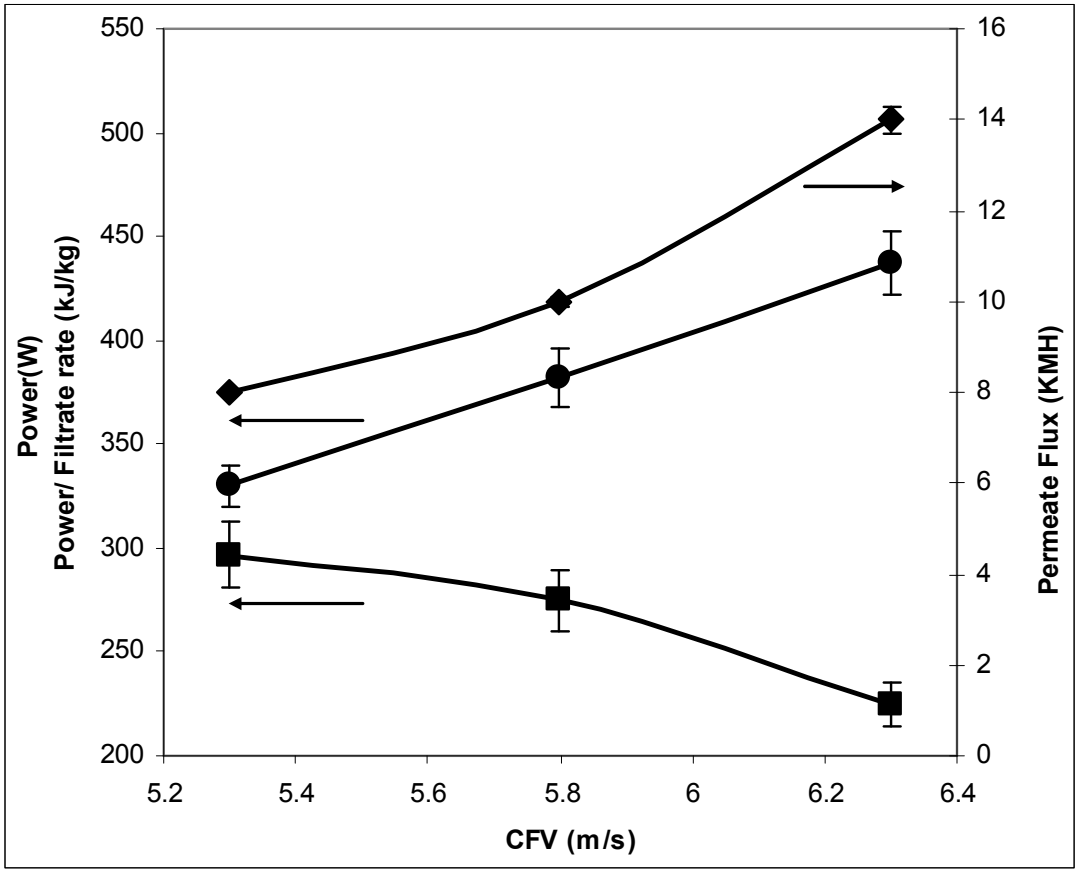


**Figure 19. Effect of CFV on power consumed (●), Permeate Flux (◆) and P/F ratio (■) during CFM of 8x retentate at 137.9 kPa**

can be maintained consistently through out the experiment, taking into account the increase in viscosity/density. The optimum CFV was 6.3 $ms^{-1}$ for our system. Although we could achieve 7.2 $ms^{-1}$ CFV at the startup, this was impossible to maintain at higher CF as the viscosity increased.

### 3.4.3. Effect of UTMP on energy consumption

As shown earlier, permeate flux increases initially (at lower concentrations) with an increase in UTMP. Since UTMP is not directly dependent on the cross flow rate or concentration factor, the power consumption should remain constant at any given UTMP, and changes only with CFV and concentration. The energy consumed (kJ/kg) decreased initially with an increase in UTMP because 100% increase in UTMP from 68.9 to 137.9 kPa resulted in an increase of filtrate rate by 22% from 51 to 62 KMH and reduced the energy consumption from 29 to 24 kJ/kg for skim milk (Figure 20). When dealing with a heterogeneous solution like milk, it is inevitable that using higher UTMPs would lead to increased fouling with time. This trend was observed at higher CF where increase in UTMP resulted in severe fouling and lower permeate flux (filtrate rate). This transition of drop of flux starts at CF = 6 (Figure 21) and is clearly noticeable at CF = 8 (Figure 22) where twice the UTMP resulted in half the filtrate rate (14.0 to 8.0 KMH) and almost twice the energy consumption (147 to 296 kJ/kg). However, the total time required (and cumulative energy requirement) to achieve CF of 8x was 40% lower at higher UTMP. It has been suggested that operating microfiltration at very high UTMP might shorten the life of the membrane (Lo et al., 1997). Based on the shortest time required to achieve CF = 8, it would be our recommendation to use higher UTMP, the issue of life of membrane notwithstanding.

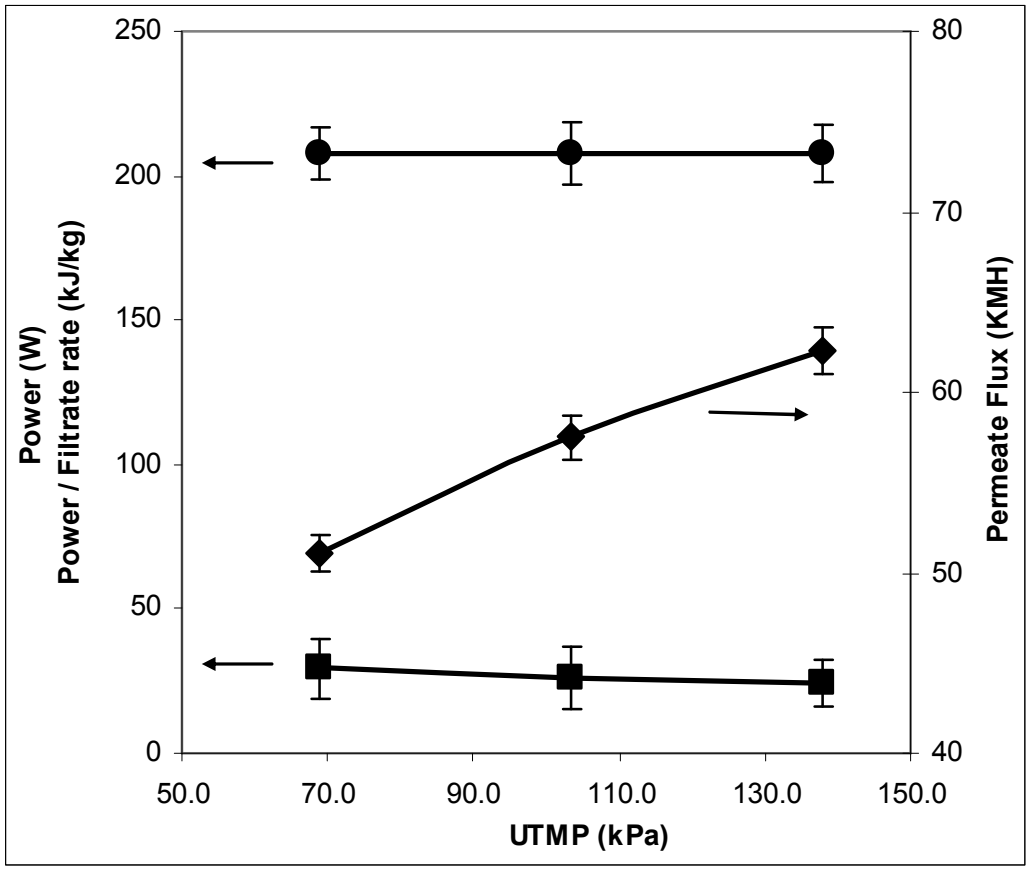


**Figure 20. Effect of UTMP on power consumed (●), Permeate Flux (◆) and P/F ratio (■) during CFM of skim milk at 5.3 $ms^{-1}$**

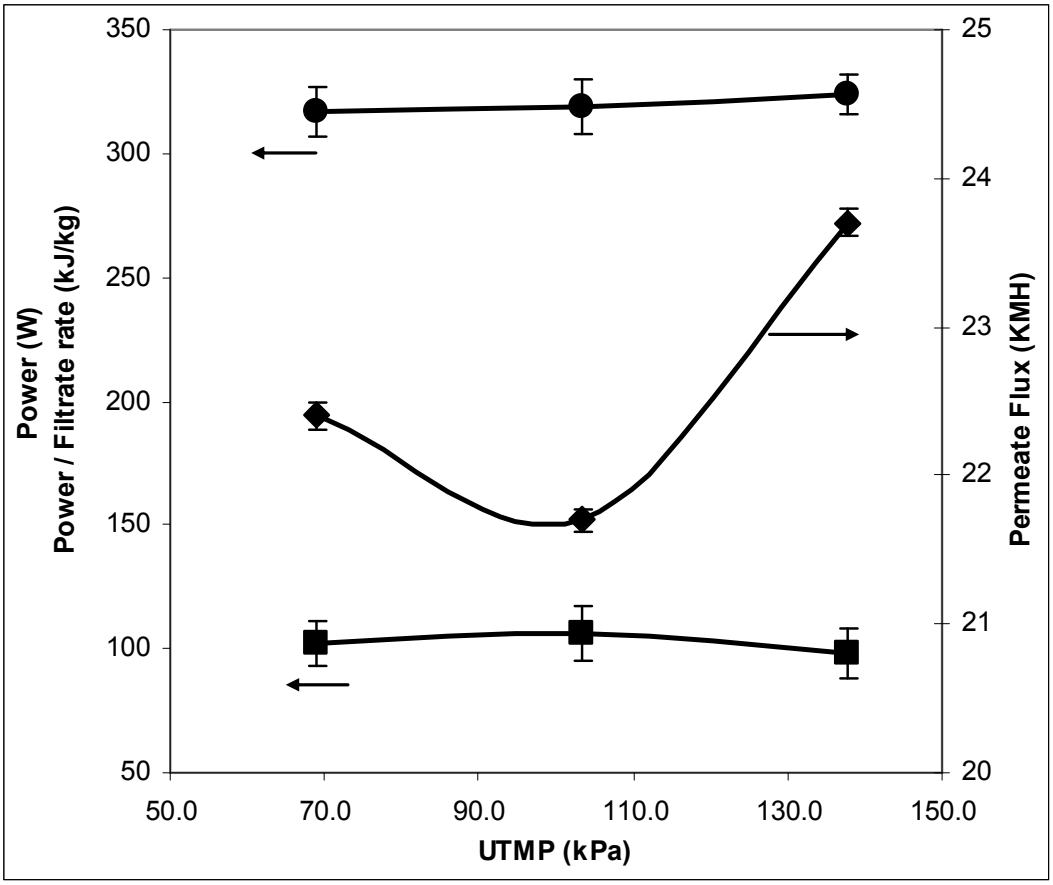


**Figure 21. Effect of UTMP on power consumed (●), Permeate Flux (◆) and P/F ratio (■) during CFM of 6x retentate at 5.3 $ms^{-1}$**

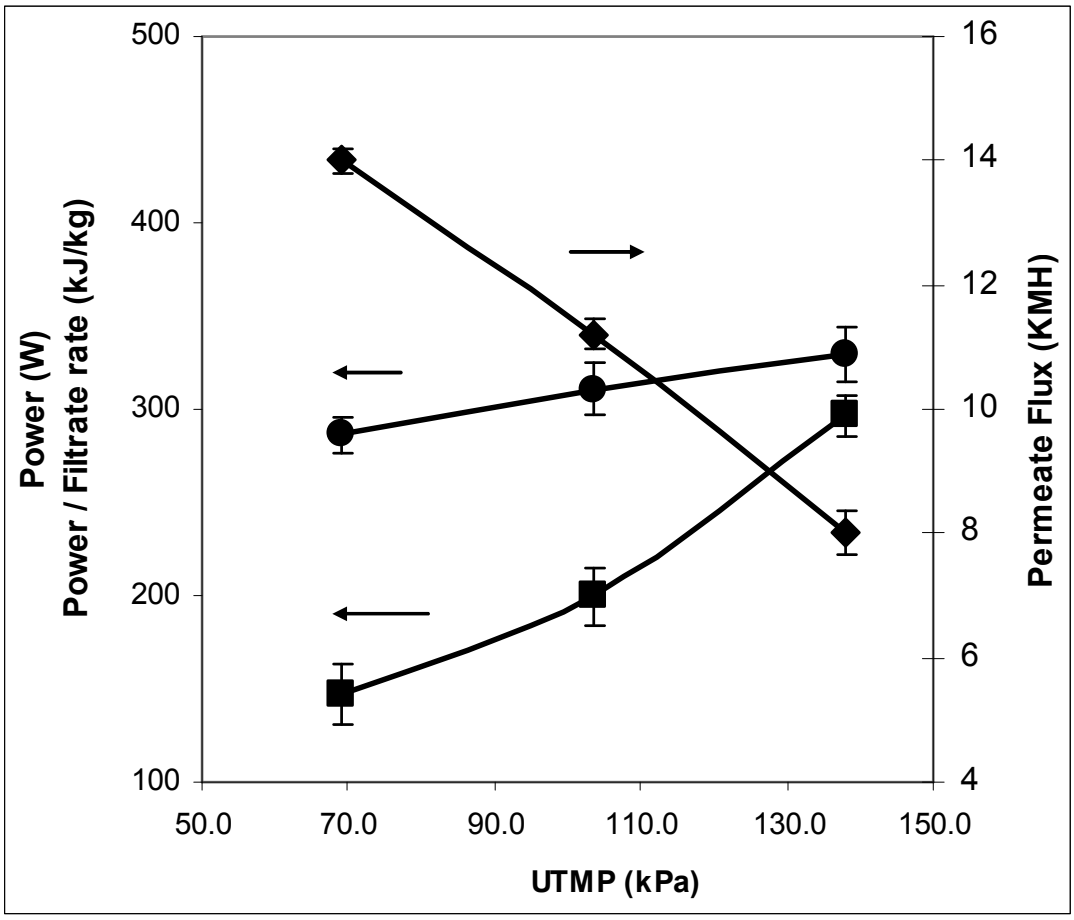


**Figure 22. Effect of UTMP on power consumed (●), Permeate Flux (◆) and P/F ratio (■) during CFM of 8x retentate at 5.3 $ms^{-1}$**

## 3.5. CONCLUSIONS

Microfiltration fills a need of the dairy industry for fractionating casein and serum proteins. Membrane processing of fluid milk permits harvesting of specific milk components without imparting a phase change, as is typical of evaporation, or using an enzyme, commonly done in most cheese making techniques. During CFM of skim milk, retentate behavior may change from Newtonian turbulent flow to pseudo plastic laminar flow at CF between 6 and 8. Almost 90 % drop in permeate flux was observed as the skim milk is selectively concentrated to 8x, while the power consumed on filtering every kg of permeate increased 12-fold. Higher flow rates resulted in higher permeate flux but also higher power consumption for skim milk. Change in power consumed per kg of permeate removed was insignificant up to CF of 4x when the flow was turbulent. For CF> 6x, an increase in flow rate reduced power consumption for every kg of permeate filtered. High UTMP was beneficial initially for CF up to 4x, as higher permeate flux was obtained with higher UTMPs. But at 6x and 8x, higher UTMP caused severe fouling and higher instantaneous power consumption, as the pressure drop across the membrane increased and permeate flux decreased. Therefore, more power was consumed for every kg of permeate filtered at 6 to 8x at higher UTMPs. Also at higher UTMPs, the permeate flux dropped precipitously around 8x, which limited the performance of CFM system. It would therefore be recommended to use the combination of high CFV and high UTMP to minimize energy consumed to manufacture highly concentrated retentates (8x), when running in batch modes. In continuous mode, a combination of high CFV and low UTMP will allow most economical cross flow microfiltration of skim milk.

## 3.6. REFERENCES


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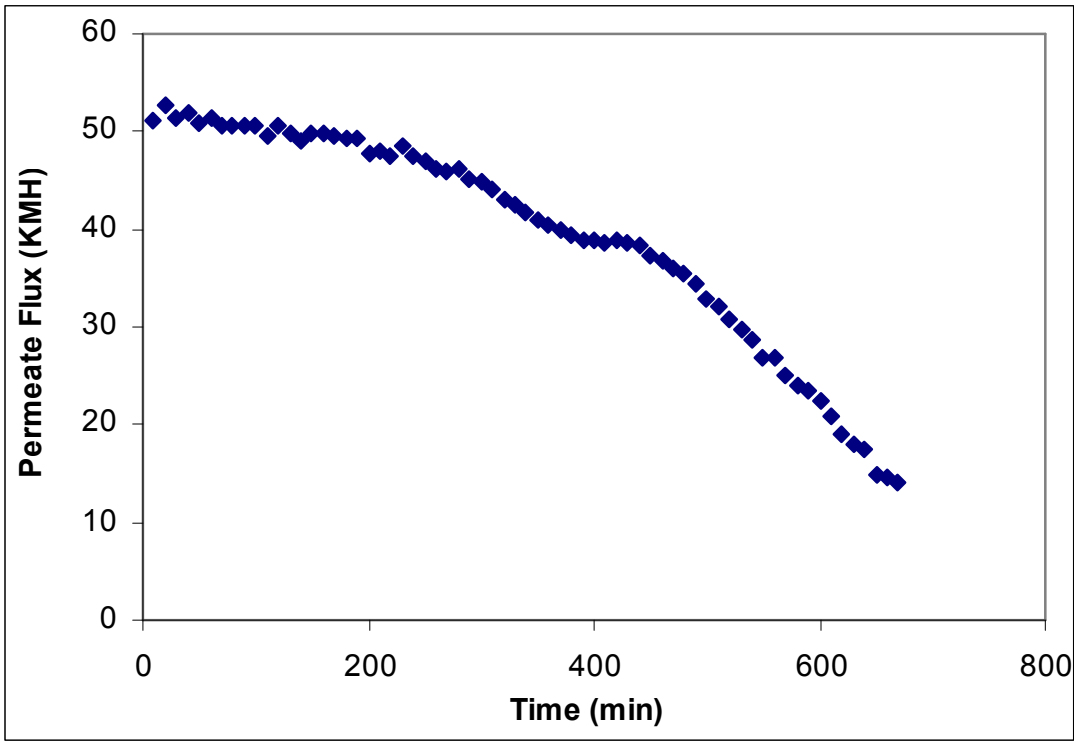
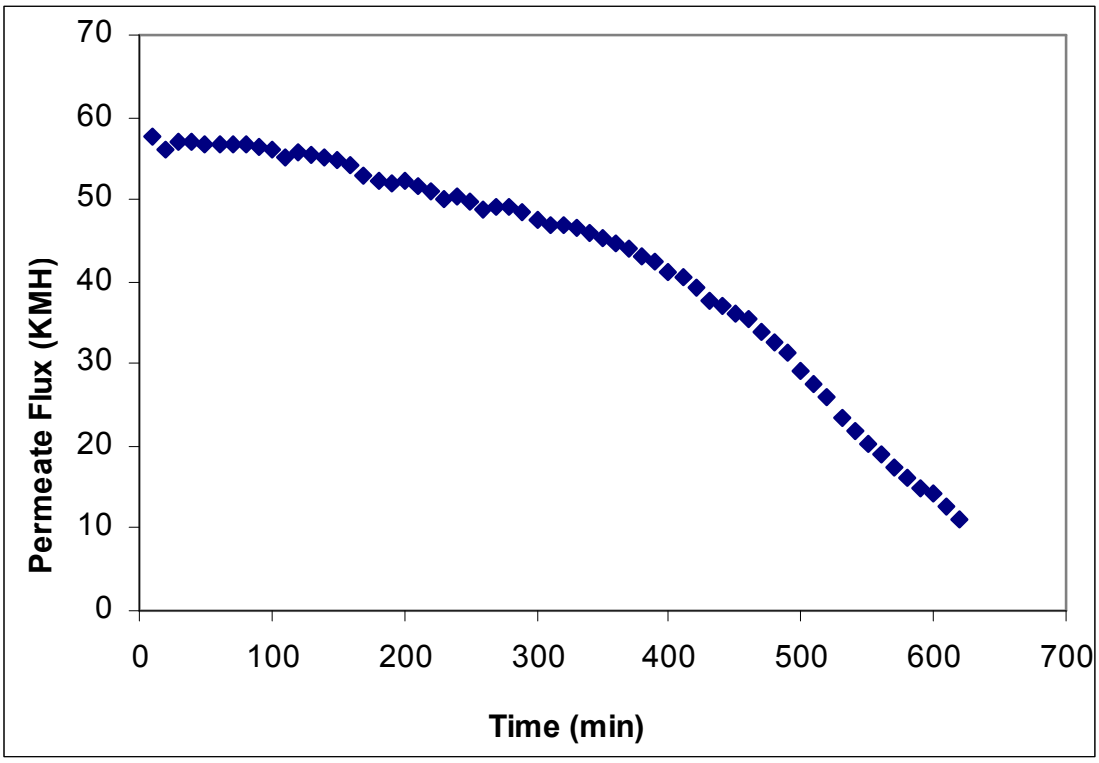
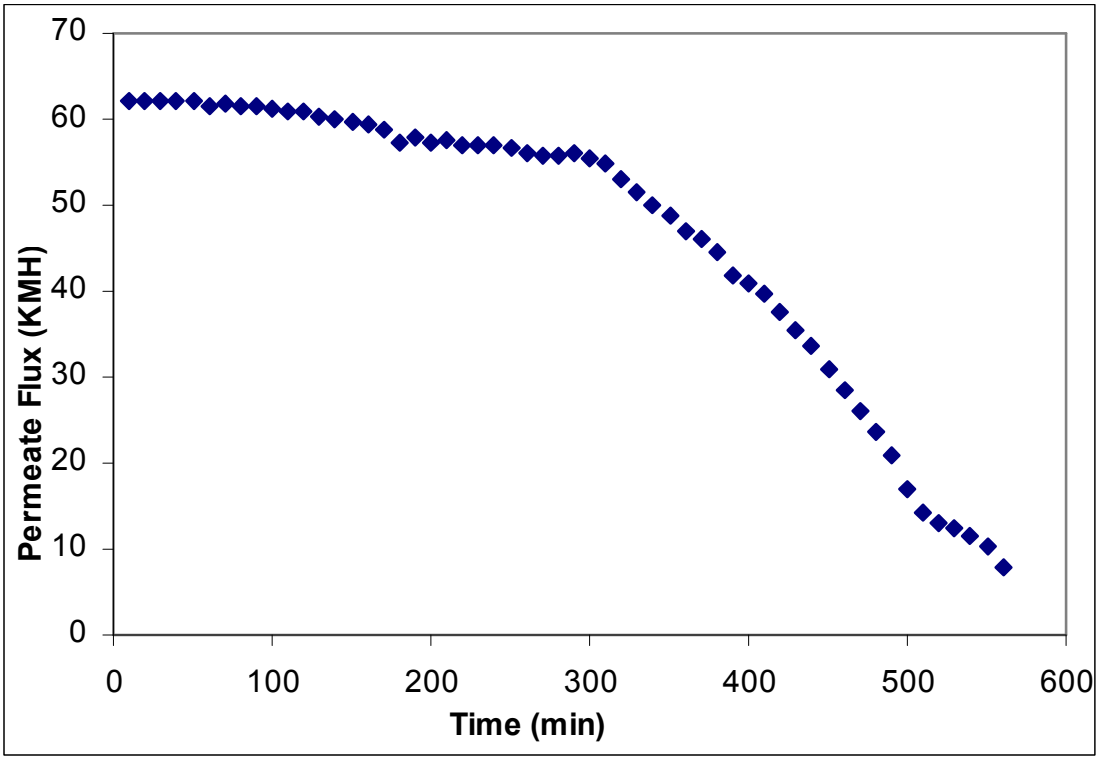
***APPENDIX 1.A.*** **Permeate Flux as a function of time for 5.3 $ms^{-1}$ and 68.9 kPa at pH 6.50**



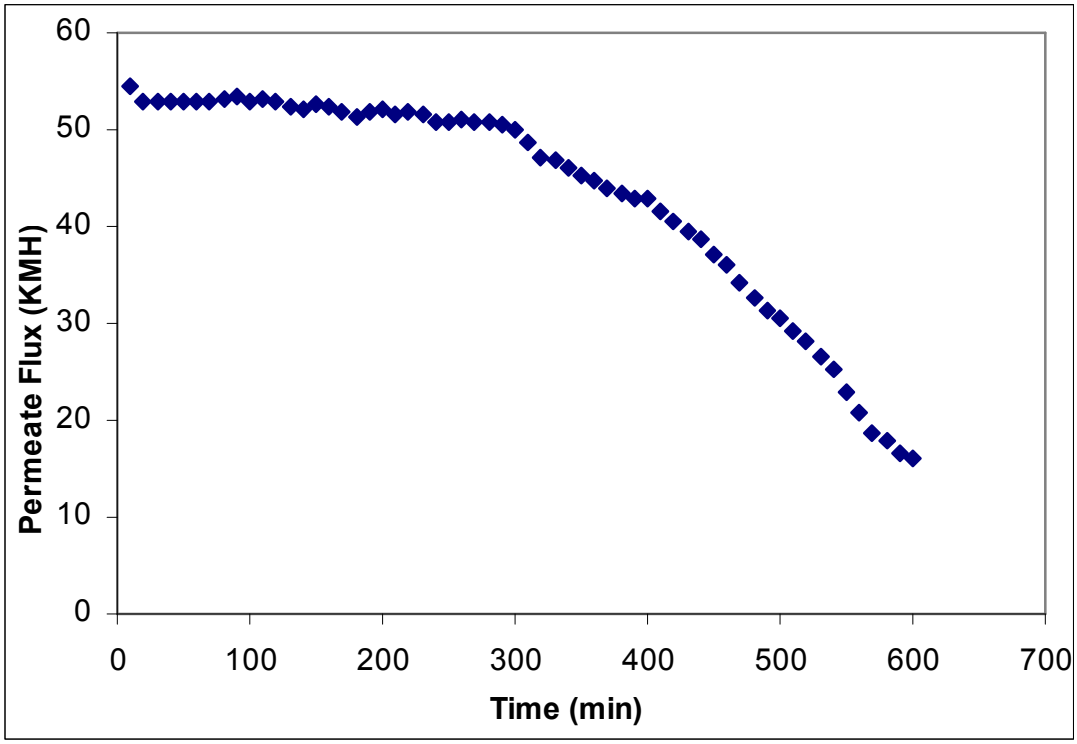
***APPENDIX 1.B.*** **Permeate Flux as a function of time for 5.3 $ms^{-1}$ and 103.4 kPa at pH 6.50**



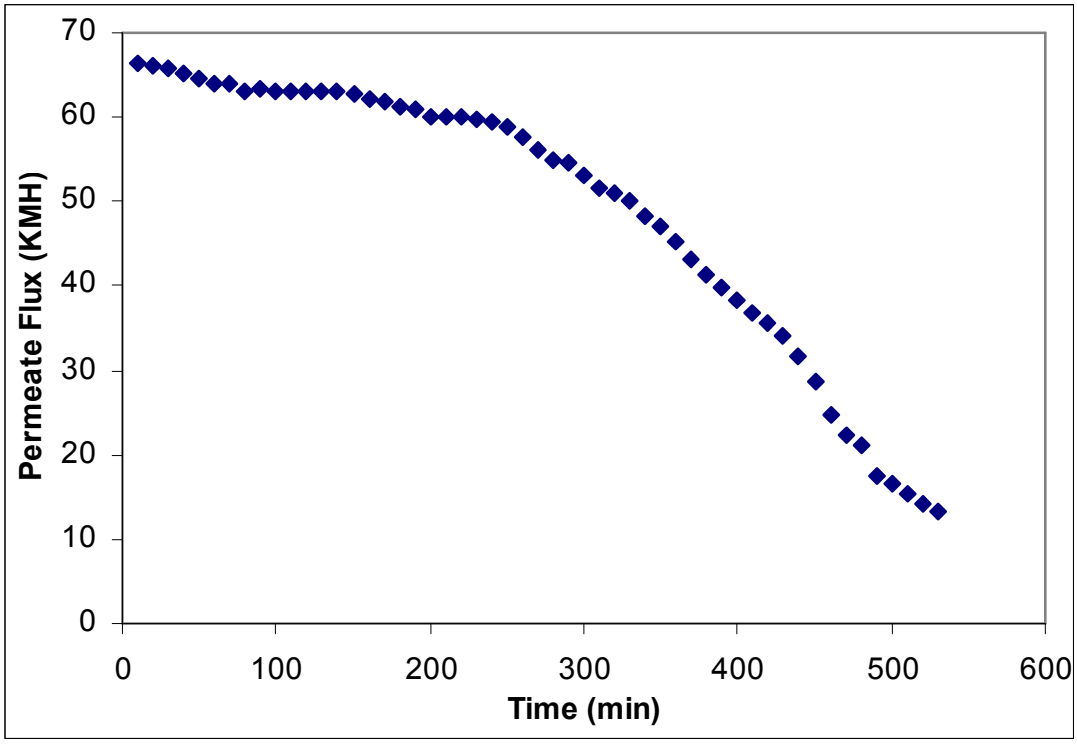
*APPENDIX 1.C.* **Permeate Flux as a function of time for 5.3 $ms^{-1}$ and 137.9 kPa at pH 6.50**



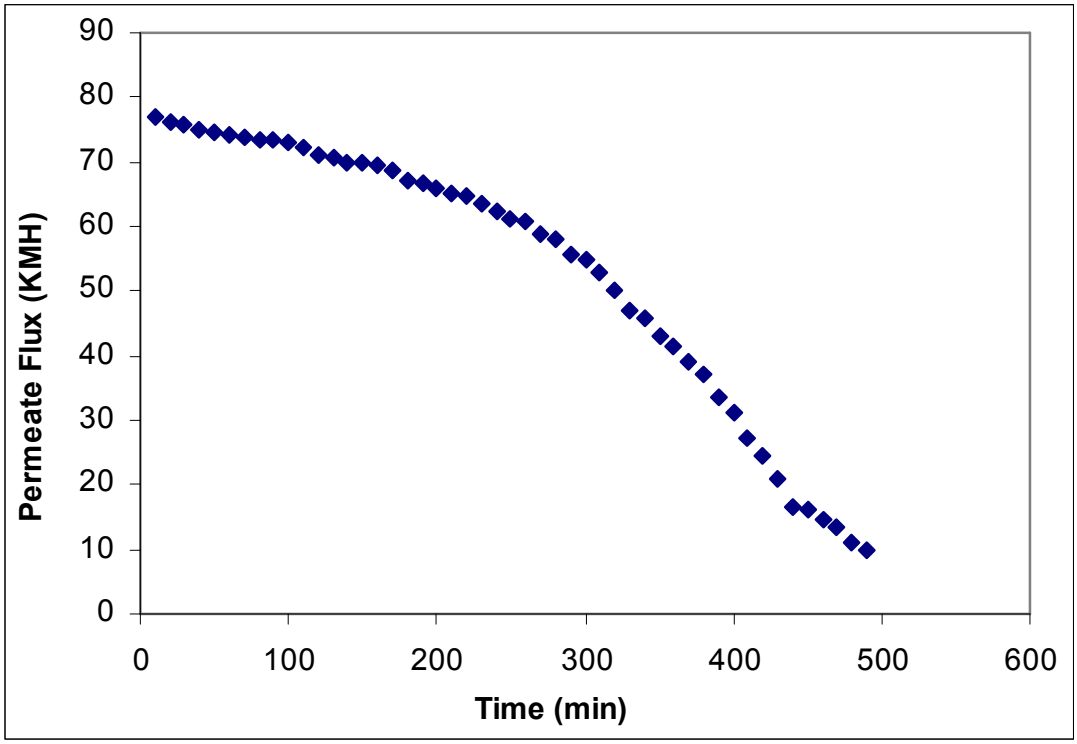
***APPENDIX 1.D.*** **Permeate Flux as a function of time for 5.8 $ms^{-1}$ and 68.9 kPa at pH 6.50**



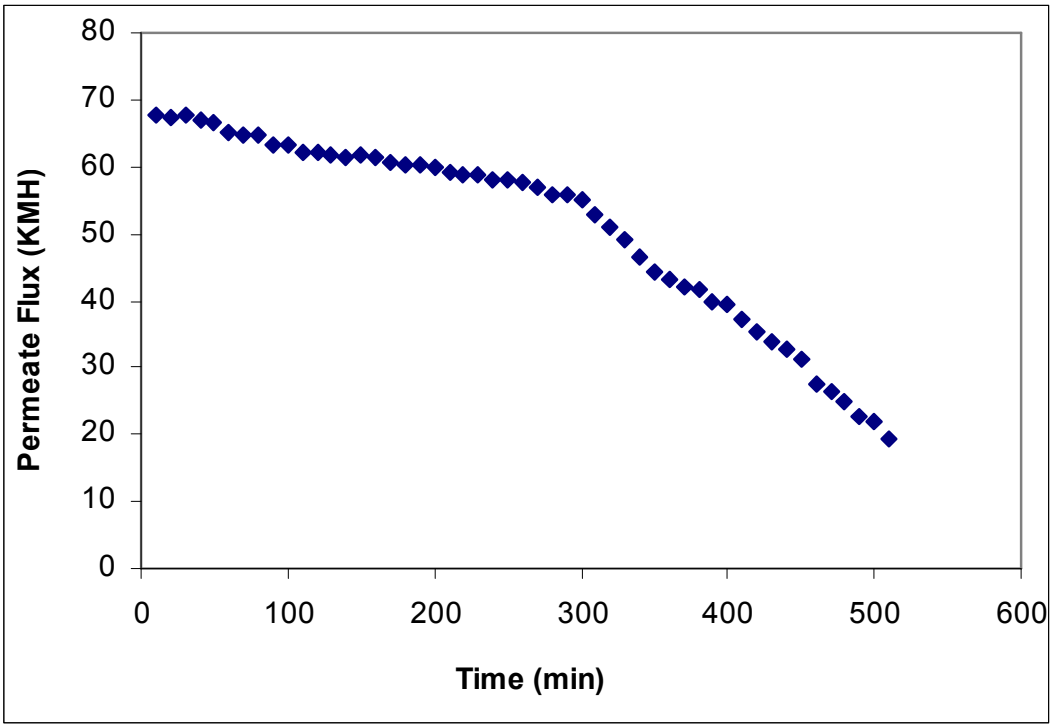
*APPENDIX 1.E.* **Permeate Flux as a function of time for 5.8 $ms^{-1}$ and 103.4 kPa at pH 6.50**



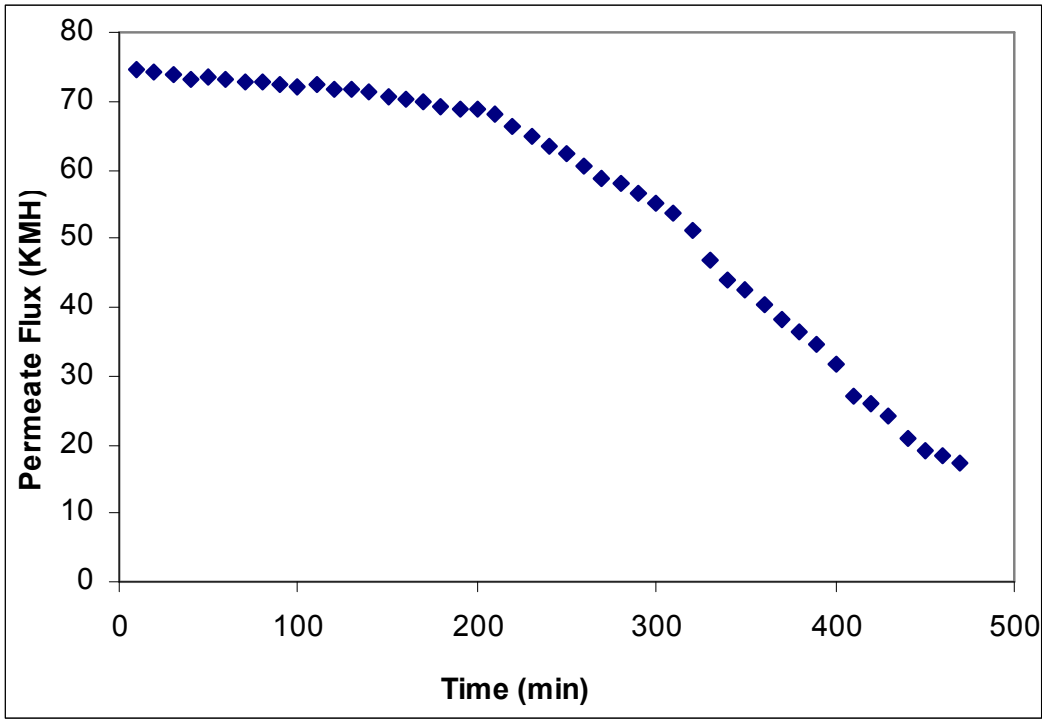
***APPENDIX 1.F.*** **Permeate Flux as a function of time for 5.8 $ms^{-1}$ and 137.9 kPa at pH 6.50**



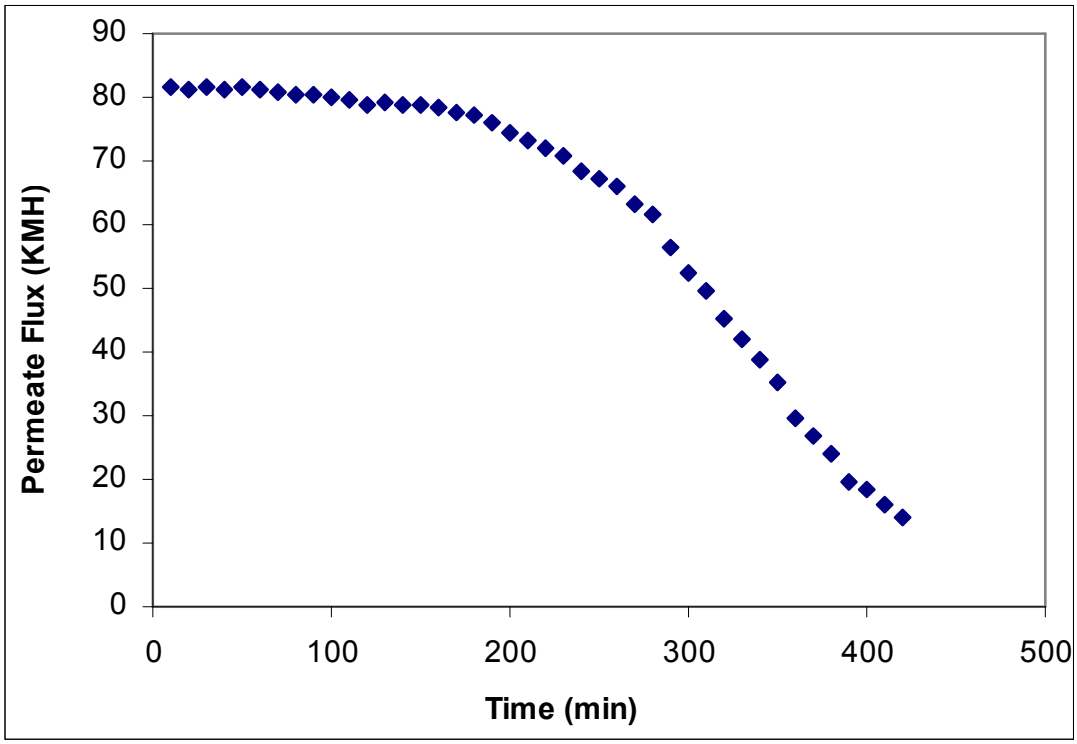
***APPENDIX 1.G.*** **Permeate Flux as a function of time for 6.3 $ms^{-1}$ and 68.9 kPa at pH 6.50**



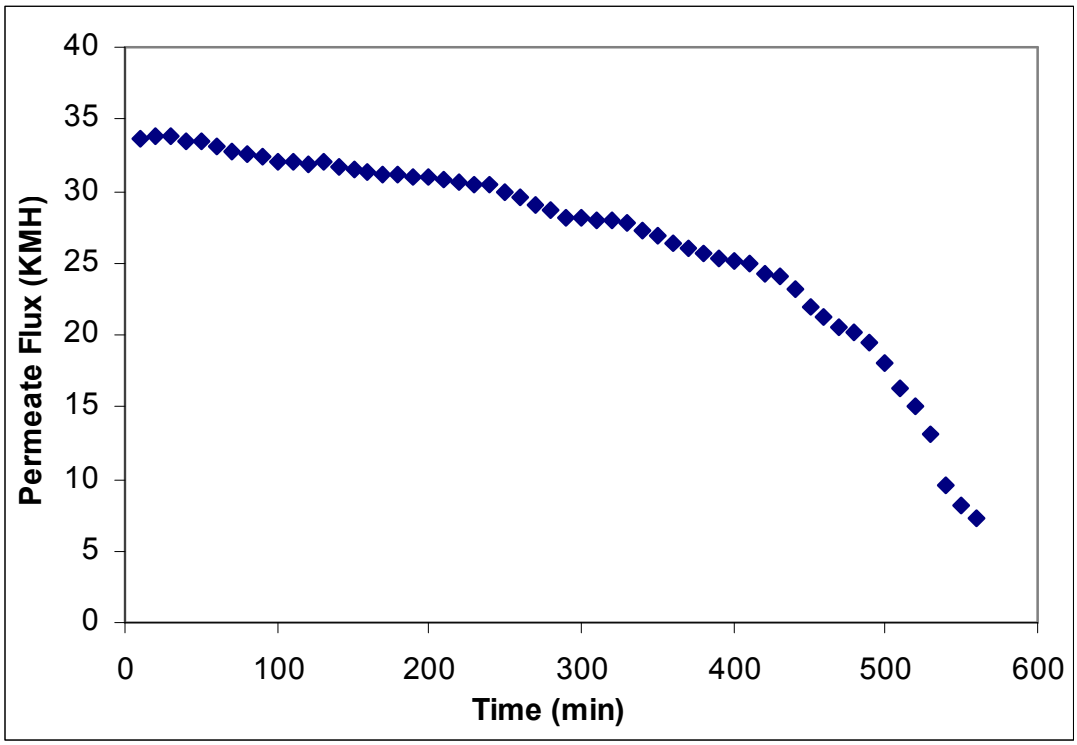
***APPENDIX 1.H.*** **Permeate Flux as a function of time for 6.3 $ms^{-1}$ and 103.4 kPa at pH 6.50**



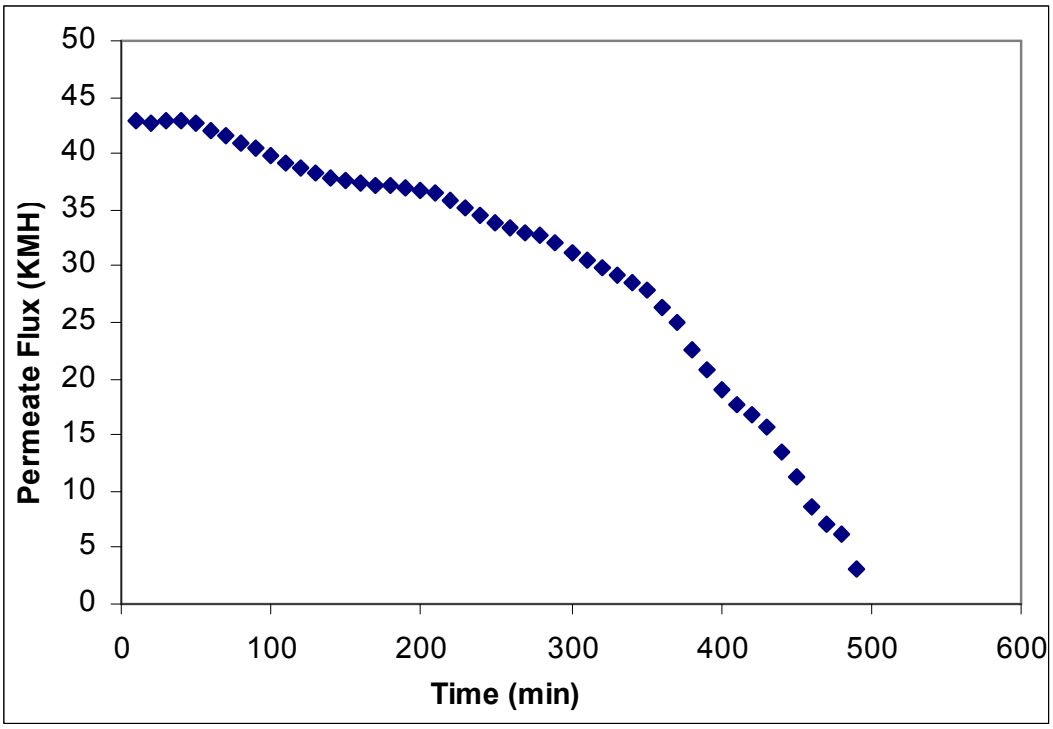
***APPENDIX 1.J.*** **Permeate Flux as a function of time for 6.3 $ms^{-1}$ and 137.9 kPa at pH 6.50**



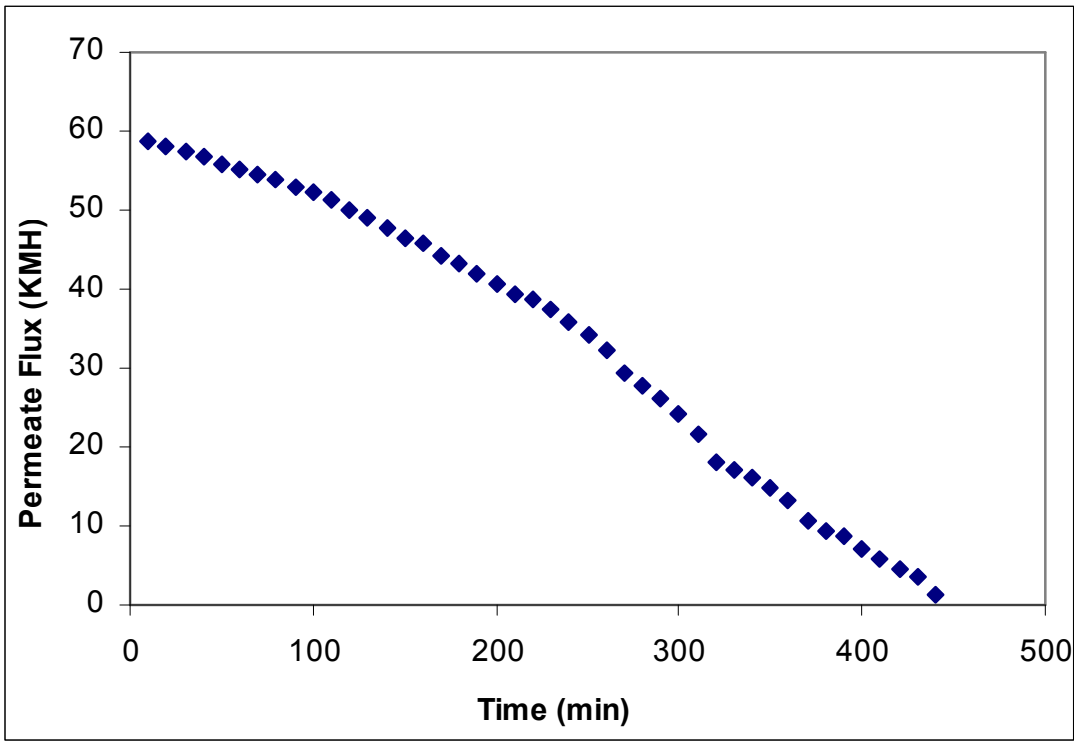
***APPENDIX 2.K.*** **Permeate Flux as a function of time for 5.3 $ms^{-1}$ and 68.9 kPa at pH 6.00**



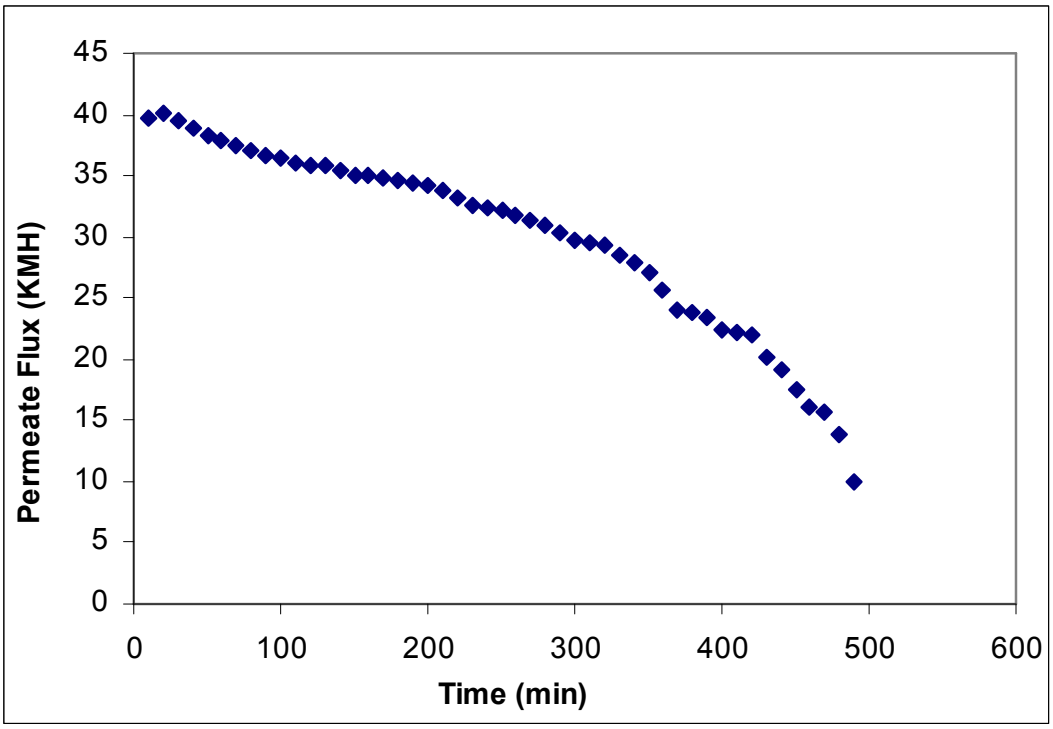
*APPENDIX 2.L.* **Permeate Flux as a function of time for 5.3 $ms^{-1}$ and 103.4 kPa at pH 6.00**



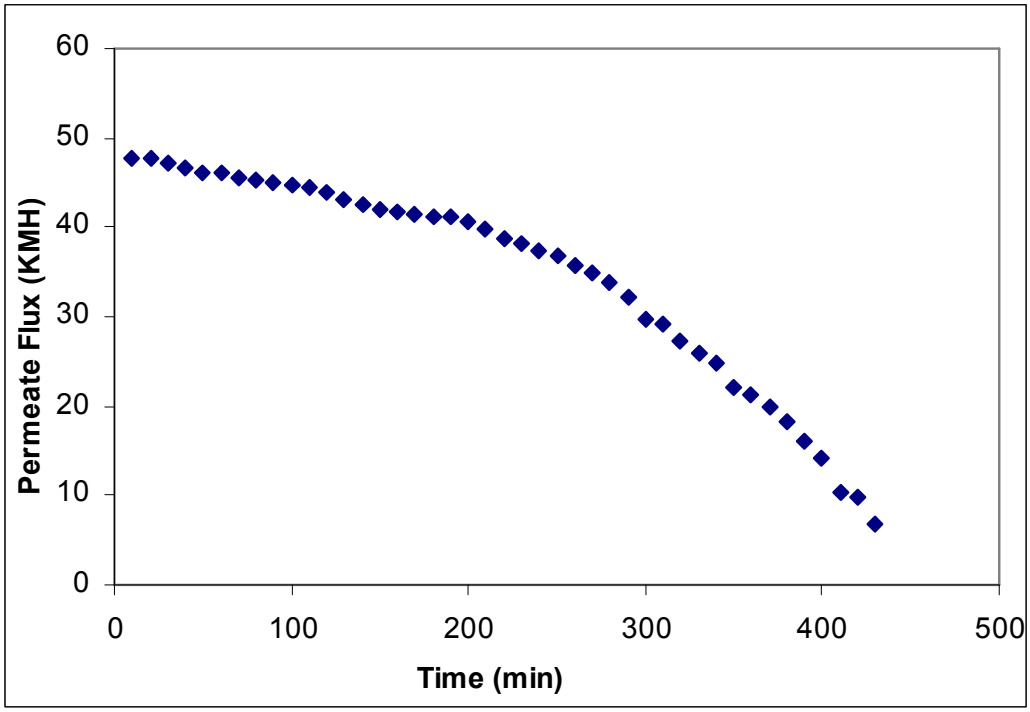
***APPENDIX 2.M.*** **Permeate Flux as a function of time for 5.3 $ms^{-1}$ and 137.9 kPa at pH 6.00**



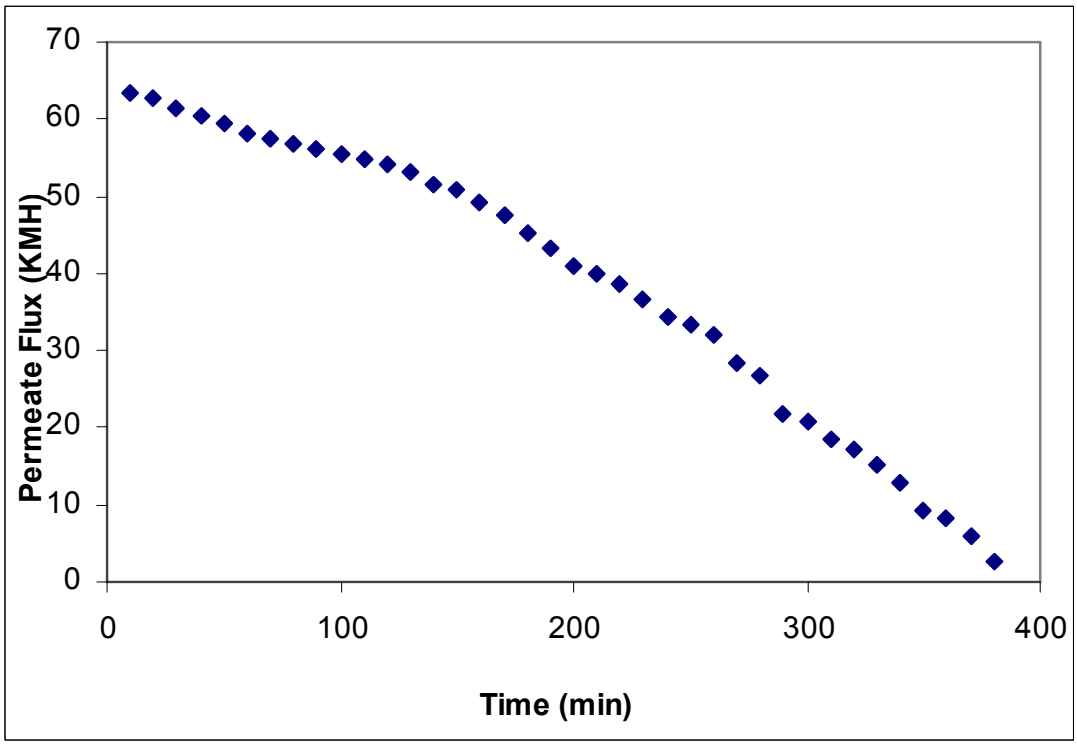
***APPENDIX 2.N.*** **Permeate Flux as a function of time for 5.8 $ms^{-1}$ and 68.9 kPa at pH 6.00**



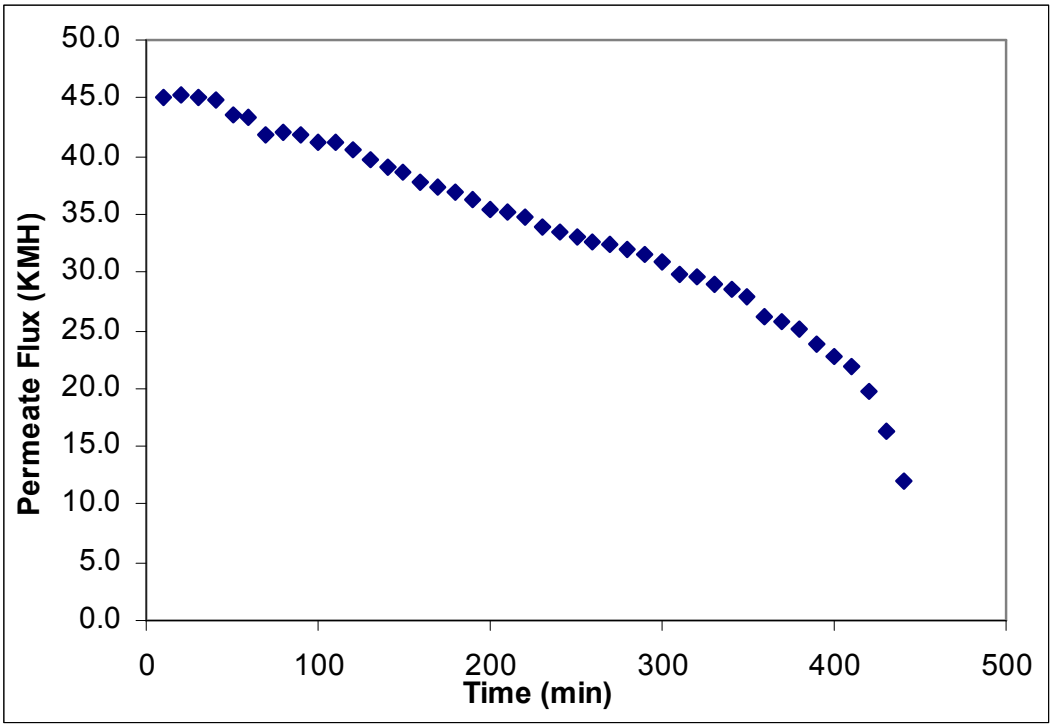
***APPENDIX 2.O.*** **Permeate Flux as a function of time for 5.8 $ms^{-1}$ and 103.4 kPa at pH 6.00**



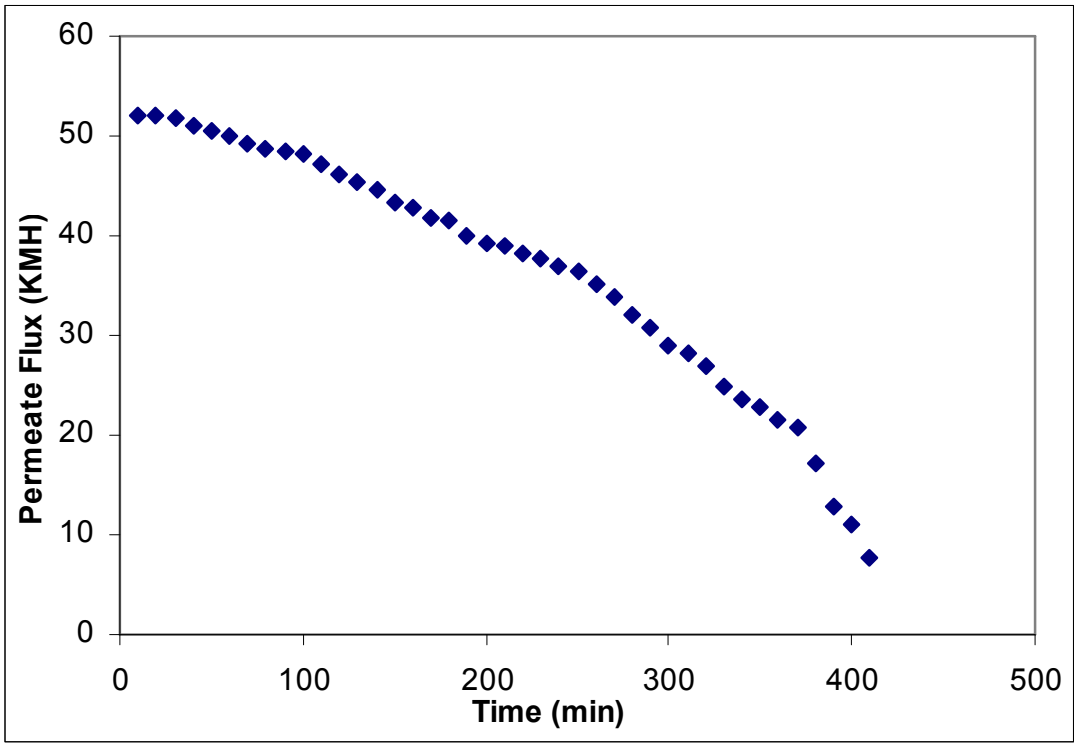
***APPENDIX 2.P.*** **Permeate Flux as a function of time for 5.8 $ms^{-1}$ and 137.9 kPa at pH 6.00**



***APPENDIX 2.Q.*** **Permeate Flux as a function of time for 6.3 $ms^{-1}$ and 68.9 kPa at pH 6.00**



***APPENDIX 2.R.*** **Permeate Flux as a function of time for 6.3 $ms^{-1}$ and 103.4 kPa at pH 6.00**



***APPENDIX 2.S.*** **Permeate Flux as a function of time for 6.3 $ms^{-1}$ and 137.9 kPa at pH 6.00**

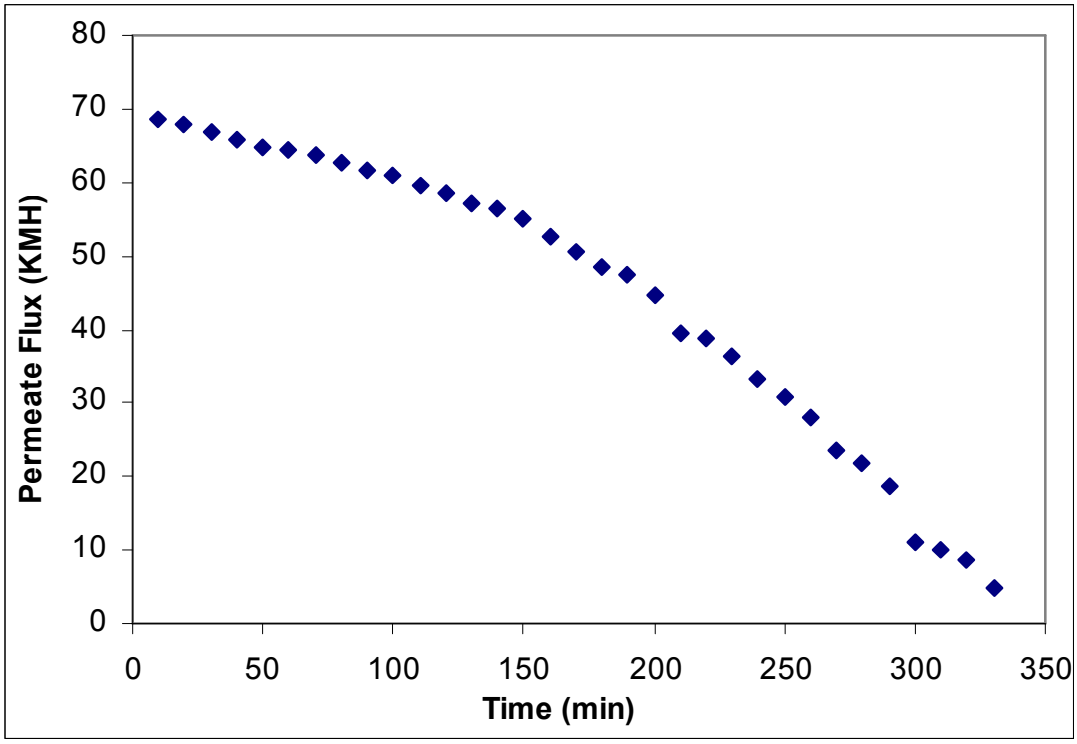


**Table 6 Effect of CFV and UTMP on permeate flux at each CF during CFM of Skim milk at pH = 6.50**

| | Permeate Flux ($kg.m^{-2}.h^{-1}$) [a] | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| CFV x UTMP | 5.3 $ms^{-1}$ 68.9kPa | 5.3 $ms^{-1}$ 103.4kPa | 5.3 $ms^{-1}$ 137.9kPa | 5.8 $ms^{-1}$ 68.9kPa | 5.8 $ms^{-1}$ 103.4kPa | 5.8 $ms^{-1}$ 137.9kPa | 6.3 $ms^{-1}$ 68.9kPa | 6.3 $ms^{-1}$ 103.4kPa | 6.3 $ms^{-1}$ 137.9kPa | Variable for P – value at each CF | |
| CF | | | | | | | | | | UTMP P – value | CFV P - value |
| 1 | 51.1 | 57.5 [b] | 62.2 | 54.4 | 66.5 | 76.9 | 67.6 | 74.6 [b] | 81.7 [b] | 0.005 | 0.007 |
| 2 | 44.0 | 49.1 | 56.8 | 50.5 | 59.5 | 65.3 | 58.0 | 68.0 [b] | 76.0 | 0.000 | 0.001 |
| 3 | 38.4 | 42.5 | 49.1 | 42.8 | 50.0 | 55.7 | 49.3 | 56.4 | 66.0 | 0.001 | 0.002 |
| 4 | 32.0 | 35.4 | 40.8 | 36.0 | 39.7 [b] | 45.6 | 40.0 | 44.0 | 52.3 | 0.001 | 0.001 |
| 5 | 26.9 [b] | 29.2 | 33.6 | 30.5 | 34.0 | 37.1 | 34.0 | 36.4 | 42.0 | 0.000 | 0.000 |
| 6 | 22.4 | 21.7 | 23.7 [b] | 25.2 | 24.8 | 27.1 | 27.4 | 27.1 | 29.6 | 0.000 | 0.001 |
| 7 | 17.6 | 16.0 | 14.3 | 18.7 | 17.6 | 16.6 | 22.6 | 21.0 | 19.8 | 0.000 | 0.001 |
| 8 | 14.0 | 11.2 | 8.0 | 16.0 | 13.2 | 10.0 [b] | 19.2 | 17.2 | 14.0 | 0.000 | 0.000 |

[a] Average Permeate Flux ($n = 2$, $P < 0.05$ for values without [b])
[b] Average Permeate Flux ($0.10 > P > 0.05$)

**Table 7. Effect of CFV and UTMP on Time required to achieve CF (2 to 8) during CFM of Skim milk at pH = 6.50**

| | Time (minutes) | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| CFV x UTMP | 5.3 $ms^{-1}$ 68.9kPa | 5.3 $ms^{-1}$ 103.4kPa | 5.3 $ms^{-1}$ 137.9kPa | 5.8 $ms^{-1}$ 68.9kPa | 5.8 $ms^{-1}$ 103.4kPa | 5.8 $ms^{-1}$ 137.9kPa | 6.3 $ms^{-1}$ 68.9kPa | 6.3 $ms^{-1}$ 103.4kPa | 6.3 $ms^{-1}$ 137.9kPa |
| CF | | | | | | | | | |
| *1* | -- | -- | -- | -- | -- | -- | -- | -- | -- |
| *2* | 316 | 282 | 252 | 286 | 238 | 210 | 238 | 210 | 190 |
| *3* | 438 | 392 | 346 | 394 | 330 | 292 | 332 | 290 | 260 |
| *4* | 510 | 456 | 402 | 458 | 386 | 342 | 388 | 340 | 302 |
| *5* | 560 | 502 | 442 | 504 | 426 | 378 | 428 | 378 | 334 |
| *6* | 600 | 542 | 476 | 540 | 460 | 410 | 460 | 410 | 362 |
| *7* | 636 | 580 | 514 | 572 | 494 | 442 | 488 | 440 | 390 |
| ***8*** | **670** | **620** | **562** | **602** | **528** | **482** | **514** | **468** | **422** |

**Table 8. Ranking of all CFV and UTMP combinations on Time required (and Instantaneous Permeate Flux) at each CF (2 to 8) during CFM of Skim milk at pH = 6.50**

| | RANK [a] (Instantaneous Permeate Flux [b]) | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| CFV x UTMP | 5.3 $ms^{-1}$ 68.9kPa | 5.3 $ms^{-1}$ 103.4kPa | 5.3 $ms^{-1}$ 137.9kPa | 5.8 $ms^{-1}$ 68.9kPa | 5.8 $ms^{-1}$ 103.4kPa | 5.8 $ms^{-1}$ 137.9kPa | 6.3 $ms^{-1}$ 68.9kPa | 6.3 $ms^{-1}$ 103.4kPa | 6.3 $ms^{-1}$ 137.9kPa |
| CF | | | | | | | | | |
| *1* | -- (9) | -- (7) | -- (6) | -- (8) | -- (5) | -- (2) | -- (4) | -- (3) | -- (1) |
| *2* | 9 (9) | 8 (8) | 6 (6) | 7 (7) | 4 (4) | 2 (3) | 4 (5) | 2 (2) | 1 (1) |
| *3* | 9 (9) | 7 (8) | 6 (6) | 8 (7) | 4 (4) | 3 (3) | 5 (5) | 2 (2) | 1 (1) |
| *4* | 9 (9) | 7 (8) | 6 (4) | 8 (7) | 4 (6) | 3 (2) | 5 (5) | 2 (3) | 1 (1) |
| *5* | 9 (9) | 7 (8) | 6 (6) | 8 (7) | 4 (4) | 2 (2) | 5 (4) | 2 (3) | 1 (1) |
| *6* | 9 (8) | 8 (9) | 6 (7) | 7 (5) | 4 (6) | 2 (3) | 4 (2) | 2 (3) | 1 (1) |
| *7* | 9 (5) | 8 (8) | 6 (9) | 7 (4) | 5 (5) | 3 (7) | 4 (1) | 2 (2) | 1 (3) |
| *8* | 9 (4) | 8 (7) | 6 (9) | 7 (3) | 5 (6) | 3 (8) | 4 (1) | 2 (2) | 1 (4) |

[a] Based on total Time required to achieve each CF

[b] Rank based on Average instantaneous Permeate Flux at each CF

**Table 9. Effect of CFV and UTMP on permeate flux at each CF during CFM of Skim milk at pH = 6.00**

| | Permeate Flux ($kg.m^{-2}.h^{-1}$) [a] | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| CFV x UTMP | 5.3 $ms^{-1}$ 68.9kPa | 5.3 $ms^{-1}$ 103.4kPa | 5.3 $ms^{-1}$ 137.9kPa | 5.8 $ms^{-1}$ 68.9kPa | 5.8 $ms^{-1}$ 103.4kPa | 5.8 $ms^{-1}$ 137.9kPa | 6.3 $ms^{-1}$ 68.9kPa | 6.3 $ms^{-1}$ 103.4kPa | 6.3 $ms^{-1}$ 137.9kPa | Variable for P – value at each CF | |
| CF | | | | | | | | | | UTMP P – value | CFV P - value |
| 1 | 33.7 | 43.0 | 58.8 | 39.8 | 47.6 [b] | 63.4 | 45.1 | 52.0 | 68.6 | 0.000 | 0.000 |
| 2 | 29.5 | 36.5 | 45.8 | 32.6 | 41.2 | 50.7 | 35.5 | 41.5 | 57.2 [b] | 0.001 | 0.023 |
| 3 | 26.4 | 32.0 | 37.3 | 29.3 | 35.7 | 40.9 | 31.6 | 36.5 | 48.4 | 0.004 | 0.037 |
| 4 | 24.3 | 28.5 [b] | 29.5 | 26.1 | 29.8 | 34.2 | 29.0 | 30.8 | 39.6 | 0.021 | 0.062 |
| 5 | 22.0 | 25.1 | 24.3 | 24.0 | 26.0 | 28.5 | 26.1 | 26.9 | 33.1 | 0.081 | 0.068 |
| 6 | 20.1 | 20.7 | 18.0 [b] | 22.3 | 22.0 | 21.7 | 23.8 | 23.6 | 28.2 | 0.933 | 0.070 |
| 7 | 18.0 | 16.8 | 14.8 | 20.2 | 20.0 | 18.4 | 22.8 | 21.5 | 23.6 | 0.462 | 0.011 |
| 8 | 15.0 | 13.5 | 10.6 | 17.6 | 16.0 | 15.3 | 19.7 | 17.3 | 18.6 | 0.100 | 0.010 |
| 9 | 9.6 | 8.6 | 7.0 | 13.9 | 10.5 | 9.1 [b] | 16.3 | 12.9 | 11.1 | 0.008 | 0.004 |
| 10 | 7.2 | 3.2 [b] | 1.3 | 10.0 | 6.8 | 2.6 | 12.0 | 7.8 | 4.9 [b] | 0.000 | 0.002 |

[a] Average Permeate Flux ($n = 2$, $P < 0.05$ for values without [b])

[b] Average Permeate Flux ($0.10 > P > 0.05$)

**Table 10. Effect of CFV and UTMP on Time required to achieve CF (2 to 10) during CFM of Skim milk at pH = 6.00**

| | Time (minutes) | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| CFV x UTMP | 5.3 $ms^{-1}$ 68.9kPa | 5.3 $ms^{-1}$ 103.4kPa | 5.3 $ms^{-1}$ 137.9kPa | 5.8 $ms^{-1}$ 68.9kPa | 5.8 $ms^{-1}$ 103.4kPa | 5.8 $ms^{-1}$ 137.9kPa | 6.3 $ms^{-1}$ 68.9kPa | 6.3 $ms^{-1}$ 103.4kPa | 6.3 $ms^{-1}$ 137.9kPa |
| CF | | | | | | | | | |
| *1* | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 | 0 |
| *2* | 264 | 210 | 160 | 230 | 188 | 146 | 206 | 178 | 132 |
| *3* | 364 | 292 | 226 | 320 | 260 | 206 | 288 | 250 | 184 |
| *4* | 418 | 338 | 268 | 370 | 302 | 242 | 334 | 292 | 216 |
| *5* | 454 | 370 | 298 | 404 | 332 | 268 | 364 | 320 | 238 |
| *6* | 480 | 394 | 324 | 428 | 356 | 290 | 386 | 342 | 256 |
| *7* | 500 | 416 | 348 | 446 | 374 | 310 | 404 | 360 | 272 |
| *8* | 518 | 436 | 372 | 462 | 390 | 328 | 418 | 376 | 286 |
| *9* | 536 | 456 | 398 | 476 | 408 | 346 | 430 | 392 | 302 |
| ***10*** | **558** | **488** | **442** | **492** | **430** | **378** | **444** | **410** | **326** |

**Table 11. Ranking of all CFV and UTMP combinations on Time required (and Instantaneous Permeate Flux) at each CF (2 to 10) during CFM of Skim milk at pH = 6.00**

| | RANK [a] (Instantaneous Permeate Flux [b]) | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| CFV x UTMP | 5.3 $ms^{-1}$ 68.9kPa | 5.3 $ms^{-1}$ 103.4kPa | 5.3 $ms^{-1}$ 137.9kPa | 5.8 $ms^{-1}$ 68.9kPa | 5.8 $ms^{-1}$ 103.4kPa | 5.8 $ms^{-1}$ 137.9kPa | 6.3 $ms^{-1}$ 68.9kPa | 6.3 $ms^{-1}$ 103.4kPa | 6.3 $ms^{-1}$ 137.9kPa |
| CF | | | | | | | | | |
| *1* | -- (9) | -- (7) | -- (3) | -- (8) | -- (5) | -- (2) | -- (6) | -- (4) | -- (1) |
| *2* | 9 (9) | 7 (6) | 3 (3) | 8 (8) | 5 (5) | 2 (2) | 6 (7) | 4 (4) | 1 (1) |
| *3* | 9 (9) | 7 (6) | 3 (3) | 8 (8) | 5 (5) | 2 (2) | 6 (7) | 4 (4) | 1 (1) |
| *4* | 9 (9) | 7 (7) | 3 (5) | 8 (8) | 5 (4) | 2 (2) | 6 (6) | 4 (3) | 1 (1) |
| *5* | 9 (9) | 7 (6) | 3 (7) | 8 (8) | 5 (5) | 2 (2) | 6 (4) | 4 (3) | 1 (1) |
| *6* | 9 (8) | 7 (7) | 3 (9) | 8 (4) | 5 (5) | 2 (6) | 6 (2) | 4 (3) | 1 (1) |
| *7* | 9 (7) | 7 (8) | 3 (9) | 8 (4) | 5 (5) | 2 (6) | 6 (2) | 4 (3) | 1 (1) |
| *8* | 9 (7) | 7 (8) | 3 (9) | 8 (3) | 5 (5) | 2 (6) | 6 (1) | 4 (4) | 1 (2) |
| *9* | 9 (6) | 7 (8) | 4 (9) | 8 (2) | 5 (5) | 2 (7) | 6 (1) | 3 (3) | 1 (4) |
| *10* | 9 (4) | 7 (7) | 5 (9) | 8 (2) | 4 (5) | 2 (8) | 6 (1) | 3 (3) | 1 (6) |

[a] Based on total Time required to achieve each CF

[b] Rank based on Average instantaneous Permeate Flux at each CF