A process efficiency assessment of serum protein removal from milk using ceramic graded permeability microfiltration membrane

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ABSTRACT

Microfiltration (MF) is a well-known process that can be used in the dairy industry to separate caseins from serum proteins (SP) in skim milk using membranes with a pore diameter of 0.1 μm . Graded permeability ceramic membranes have been studied widely as means of improving milk fractionation by overcoming problems encountered with other MF membranes. The ideal operating parameters for process efficiency in terms of membrane selectivity, permeate flux, casein loss, SP transmission, energy consumption, and dilution with water remain to be determined for this membrane. Our objective was to evaluate the effects of transmembrane pressure (TMP), volumetric concentration factor (VCF), and diafiltration on overall process efficiency. Skim milk was processed using a pilot-scale MF system equipped with 0.72-m² graded permeability membranes with a pore size of 0.1 μm . In the first experiment, in full recycle mode, TMP was set at 124, 152, 179, or 207 kPa by adjusting the permeate pressure at the outlet. Whereas TMP had no significant effect on permeate and retentate composition, 152 kPa was found to be optimal for SP removal during concentration and concentration or diafiltration experiments. When VCF was increased to 3 $\times$, SP rejection coefficient increased along with energy consumption and total casein loss, whereas SP removal rate decreased. Diafiltering twice allowed an increase in total SP removal but resulted in a substantial increase in energy consumption and casein loss. It also reduced the SP removal rate by diluting permeate. The membrane surface area required for producing cheese milk by blending whole milk, cream, and MF retentate (at different VCF) was estimated for different cheese milk casein concentrations. For a given casein concentration, the same quantity of permeate and SP would be produced, but less membrane surface area would be needed at a lower retentate VCF. Mi-

crofiltration has great potential as a process of adding value to conventional cheesemaking processes, but its cost-effectiveness at a large scale remains to be demonstrated.

Key words: microfiltration, graded permeability membrane, process efficiency, energy consumption, mass balance

INTRODUCTION

Microfiltration (MF) is a pressure-driven, membrane-based process used to concentrate particles in the 0.1 to 10 μm size range and to separate them from the suspending liquid (Saboya and Maubois, 2000). The first major application of MF in the dairy industry was for the removal of bacteria from milk using a membrane having a pore size of 1.4 μm (Trouvé et al., 1991). A pore size of 0.1 μm allows the separation of casein micelles from SP of skim milk (Saboya and Maubois, 2000). The casein-enriched retentate thus obtained can be used to standardize cheese milk, whereas the clear, fat-free, and sterile serum protein (SP)-enriched permeate has a composition similar to cheese whey. Serum proteins contained in the MF permeate are functionally and nutritionally superior to SP from cheese whey, which gives it an excellent potential for production of SP concentrates or isolates (Saboya and Maubois, 2000). This process also allows the production of micellar casein concentrate (MCC), a novel dairy ingredient obtained by diafiltering the casein-enriched retentate. This high-protein ingredient can be used in the formulation of different products due to its nutritive and functional value, as well as its stability during processing.

Ceramic is a popular choice of membrane material for MF applications due to its resistance to back-pressure and steam sterilization, its narrow pore size distribution, and the high permeate flux obtainable (Zulewska et al., 2009; Fernández García and Riera Rodríguez, 2015). First-generation ceramic membranes fouled quickly, decreasing selectivity, permeate flux, and overall performance (Saboya and Maubois, 2000). To overcome this problem, Sandblom (1978) developed the uniform transmembrane pressure (UTP) system,

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whereas Alfa Laval (Kolding, Denmark) improved and patented this process a few years later (Holm et al., 1990). In this system, MF permeate is recirculated in a loop parallel to the retentate flow, which allows a lower and more UTP from the retentate inlet to the outlet and reduces membrane fouling. Although the UTP system performed well, it was not adopted widely, due mainly to high operating costs associated with the permeate recirculation pump and the difficulty of adjusting pressure drops on the permeate and retentate sides (Hu and Dickson, 2015). In recent years, several attempts were made to achieve the benefits of UTP system, but without the permeate recirculation pump (Saboya and Maubois, 2000). One promising alternative to UTP systems is the graded permeability (GP) ceramic membrane (Membralox GP, Pall Corp., East Hills, NY). With GP membranes, uniform permeate flux is obtained by incorporating a variation in the porosity of the membrane support along the length of the filtration element (Garcera and Toujas, 2002). This allows MF with less energy expenditure and simpler operation than with UTP systems (Hu and Dickson, 2015).

A study comparing UTP and GP systems for separating SP from skim milk has shown that UTP allows greater passage of SP, whereas permeate flux is greater through the GP membrane (Zulewska et al., 2009). Spiral-wound polymeric membrane (0.3 μm pore size) and Isoflux (TAMI, Nyons, France) ceramic membrane have been found less effective than UTP or GP systems (Zulewska et al., 2009; Adams and Barbano, 2013). Previous work by Zulewska and Barbano (2014) evaluated the effect of diafiltration (DF) on the efficiency of SP removal during MF of skim milk. Most studies of the performance of GP membranes have been conducted under feed-and-bleed conditions, which allow continuous production of retentate with a volumetric concentration factor (VCF) of $3\times$ by adjusting permeate and retentate removal rates during the process. Whereas this process is used widely by dairy processors, it is prone to fluctuating operating pressures and Starling flow (permeate backflow into the retentate), which increases membrane fouling and reduces the effective filtration surface area (Zulewska et al., 2009).

Efficiency can be defined as the link between results obtained and the amount of resources used. For a dairy processor, the ideal MF process would yield retentate sufficiently enriched in casein to be useful for standardizing cheese milk, plus permeate containing high-value SP and as little casein as possible would require low energy input and minimal dilution with water and would have a high permeate flux (Noble and Agrawal, 2005). To maximize process efficiency, MF should be performed at a transmembrane pressure (TMP) at

or below the critical flux defined as the mean flux at which a fouling layer begins to accumulate on the membrane (Bacchin, 2004). Raising the TMP above the critical flux increases the thickness of the fouling layer and decreases SP transmission (Gésan-Guizieu et al., 1999). Zulewska and Barbano (2014) determined that the critical flux of skim milk in a continuous feed-and-bleed process with a VCF of $3\times$ using a 0.1- μm GP membrane was 64 L/m² per hour. Under the same conditions, the maximal flux attainable (limiting flux) was estimated at 96 L/m² per hour. Sustainable flux is defined as the flux that can be maintained for a long time with minimal fouling, and is usually in between the critical and limiting fluxes (Hurt et al., 2015a). It was determined that this flux can be achieved at a TMP of 150 kPa regardless of the target protein concentration or membrane channel diameter. During MF of skim milk at a VCF of $3\times$, and using a 4-mm channel diameter GP membrane, this flux corresponds to 80 to 85% of the limiting flux (77–82 L/m² per hour; Hurt et al., 2015a). Other parameters, such as pressure drop, cross-flow velocity, operating temperature, membrane properties, type of pasteurization, feed concentration, and DF, have an influence on the efficiency of skim milk MF (Bacchin et al., 2006; Hu and Dickson, 2015).

Whereas the effects of channel diameter (Hurt et al., 2015a) and retentate protein concentration (Hurt et al., 2015b) on the limiting flux and the SP removal using GP membrane have been studied, no study has focused on attempts to perform batch filtrations at constant operating pressures. This would provide the truest assessment of GP membrane performance at different VCF and TMP. To the best of our knowledge, little attention has been devoted to processing efficiency in terms of overall mass balance under different conditions and, more importantly, to the in situ energy requirements of those conditions. The goal of our study was to characterize the performance of a 0.1- μm pore size GP membrane at different TMP and different VCF and to evaluate the effect of DF on the separation of SP from skim milk and on overall process efficiency.

MATERIALS AND METHODS

Skim Milk Preparation

Whole raw bovine milk was purchased from the bulk of a local dairy. Raw milk was heated to 45°C in a heat exchanger then separated using a centrifugal cream separator (model MP1254, Westfalia, Oelde, Germany). The cream was discarded and the skim milk (<0.05% fat) was HTST-pasteurized in a heat exchanger (APV, Goldsboro, NC) at 74°C for 16 s. The pasteurized skim milk was cooled to 4°C and stored at 4°C in a refrig-

erated tank (model 180, DeLaval, Peterborough, ON, Canada) until the experiments.

MF System

The MF system (model 393, Tetra Pak Filtration Systems, Champlin, MN) schematized in Figure 1 was equipped with 0.1- μm pore diameter Membralox ceramic GP membrane (modelr EP1940, Pall Corp.) with a surface area of 0.72 m². The tubular membranes had a configuration with 3 tubes, 19 channels each, with a channel diameter of 4 mm, in a stainless steel module that was mounted vertically in the system. Retentate inlet was located at the bottom of the membrane module and permeate was removed from the top of the module. Two feed pumps, one for nanofiltration or reverse osmosis (modelr H-25, Wanner Engineering Inc., Minneapolis, MN) and one for MF or UF (modelr LKHP-10, Alfa Laval), worked simultaneously, and one recirculation pump (modelr LKHP-10, Alfa Laval) was

used. Electromagnetic flow meters (modelr SM2001, IFM Electronic, Malvern, PA) were used to measure feed, retentate, and permeate flow rates. Pressure transmitter (modelr PF265x, IFM Electronic) locations are marked as retentate pressure at the inlet (**R_{pi}**), retentate pressure at the outlet (**R_{po}**), and permeate pressure at the outlet (**P_{po}**) in Figure 1. A heat exchanger on the feed loop and a thermocouple probe (modelr FD02, Pyromation, Fort Wayne, IN) provided temperature control.

Preliminary Operations

Prior to processing, the milk was maintained at 50°C for 1 h in a stainless steel intermediary tank (modelr DSC12336, Qualtech, Québec, QC, Canada) to establish protein and mineral balance between the soluble and colloidal phases (Jimenez-Lopez et al., 2008). The membrane storage solution (0.1% vol/vol Membra-Stor 710, Sani-Marc, Victoriaville, QC, Canada) was flushed

Figure 1. Schematic of the microfiltration system. R_{pi} = retentate pressure at the inlet; R_{po} = retentate pressure at the outlet; P_{po} = permeate pressure at the outlet; NF/RO = feed pump for high operating pressure (nanofiltration and reverse osmosis); MF/UF = feed pump for low operating pressure (microfiltration and ultrafiltration); A = feed valve; B = permeate valve. Permeate was either removed from the system and weighed on a scale during concentration or recirculated in the feed tank during the full recycle operation; Retentate was always recirculated to the feed tank.

out of the system with tap water at 25°C until the pH was neutral on the permeate and retentate sides. The MF system was then heated gradually ($<10^{\circ}\text{C}/\text{min}$) to 50°C and a caustic solution (1.95% vol/vol Membra-Chlor 310, Sani-Marc) was added to the system and recirculated for 25 min. After cleaning, the caustic solution was flushed out of the system using tap water at 50°C until the pH of the flush water was neutral. The clean water flux was measured with tap water at 50°C at a TMP of 172 kPa, with the feed valve closed, and only the MF or UF feed pump on.

Experimental Design

Three different experiments were performed in duplicate. For each experiment, one batch of milk was divided into 2 equal parts. Optimal pressure conditions were determined during an experiment conducted in full recycle mode with nonconcentrated skim milk. The pressure thus determined was used in the concentration experiment until a VCF of 3 $\times$ was obtained. Finally, the concentration/DF condition was the same as described previously except that the initial VCF 3 $\times$ retentate was diluted 1:2 with tap water and then reconcentrated to a VCF of 3 $\times$. This DF procedure was performed twice.

Determination of the Optimal TMP for SP Removal

Each replicate began with 80 kg of skim milk to flush the tap water out of the MF system with the permeate outlet closed to prevent membrane fouling (Cheryan, 1998). After the flush, the MF system was shut down and the remaining milk was flushed with manual valves. An estimated 30 kg of skim milk could not be flushed from the system. The feed tank was then filled with 100 kg of skim milk for the actual test. The system was started up and the pressure was adjusted to the lowest setting in full recycle mode. For each pressure level, Rpi and Rpo were kept constant at 310 and 103 kPa, respectively, thus providing a pressure drop (Rpi – Rpo) of 207 kPa, which is optimal for GP membrane and allows a cross-flow velocity of about 7 m/s (Adams et al., 2015). Thus, the only factor affecting the TMP was the Ppo, which was 83, 55, 28, and 0 kPa (pressure levels 1, 2, 3, and 4, respectively); average TMP were then 124, 152, 179, and 207 kPa, respectively. The Ppo was always lower than Rpo to prevent Starling flow.

Skim milk was processed at 50°C. Each pressure level was maintained for 45 min and the permeate flux was measured at 0, 15, 30, and 45 min. The pressure was allowed to stabilize for 15 min between TMP adjustments, and the milk was then recirculated for another 45 min. Permeate and retentate samples were taken after 30 min of recirculation at each pressure level. Skim

milk was sampled directly from the feed tank at the beginning of the experiment. All samples were cooled to 4°C and frozen at -18°C until analysis.

Skim Milk Concentration

Following the full recycle experiment, it was determined that a TMP of 152 kPa was optimal for SP removal and was therefore chosen for the skim milk concentration experiments. This TMP value was similar to that obtained by Hurt et al. (2015a) and corresponded to the sustainable flux estimated for skim milk MF using GP membranes. All concentration steps were carried out in batch mode to keep operating pressures as steady as possible during the process. For each replicate, water was flushed according to the same procedure as described for the full recycle experiment. After the flush, 230 kg of skim milk were added to the feed tank, bringing the total quantity of skim milk in the system to 260 kg. The MF system was started in full recycle mode until pressure conditions were established. Permeate was then removed from the system and weighed during the entire run for real-time VCF measurements. Permeate flux was measured every 15 min. Skim milk was sampled in the feed tank at the beginning of the experiment (before concentration). Permeate and retentate samples were taken at VCF of 1.5, 2, 2.5, and 3 $\times$. All samples were cooled to 4°C and frozen at -18°C until analysis.

Skim Milk Concentration and DF

Flushing and starting procedures, as well as operating pressures, were as described for the concentration experiment. For each replicate, skim milk was concentrated to a VCF of 3 $\times$, followed by 2 batch DF. For each DF, 173 kg of tap water were used, which is equivalent to the permeate weight removed during concentration. After adding the water, the MF system was put in full recycle mode for 5 min to blend the retentate, permeate, and water. Permeate was then withdrawn again to reach a VCF of 3 $\times$. Permeate flux was measured every 15 min and VCF was monitored in real-time.

Skim milk was sampled at the beginning of the experiment directly from the feed tank. Permeate and retentate samples were taken at a VCF of 3 $\times$ after initial concentration, and at the end of each DF step. All samples were cooled to 4°C and frozen at -18°C until analysis.

Cleaning Procedure

After each MF condition, the permeate outlet was closed and the system was flushed with tap water at

50°C until the retentate side water was completely clear. The fouled membrane flux was then measured using the same parameters as the initial clean water flux measurement. Thereafter, the system was sanitized with 1.95% (vol/vol) Membra-Chlor 310 solution (Sani-Marc) until the chlorine level reached 0.02% vol/vol. The caustic solution (50°C) was recirculated for 30 min and then flushed out with tap water at 50°C until the flush water pH was neutral. Ultrasil 76 (Ecolab Inc., St Paul, MN) was added (0.3% vol/vol) to the feed tank and recirculated for 30 min at 50°C. The acidic solution was flushed out with tap water at 50°C until the flush water pH was neutral. The clean water flux was measured after the cleaning procedure to confirm that it was within 10% of the initial water flux.

Chemical Analysis

All samples were thawed at 4°C for analysis. Total solids were measured by air drying in an oven (AOAC International, 2000; 990.20), whereas total nitrogen (TN), noncasein nitrogen (NCN), and NPN content were determined using Kjeldahl methods (991.20, 998.05, and 991.21, respectively; AOAC International, 2000). True protein (TP) content was calculated as $(TN - NPN) \times 6.38$, casein content was calculated as $(TN - NCN) \times 6.38$, and SP content calculated as $(NCN - NPN) \times 6.38$.

Determination of Process Efficiency

To assess the concentration and DF process efficiencies, mass balance calculations were used for SP and casein according to the method of Zulewska et al. (2009). The total weight of SP in skim milk and permeate (e.g., VCF = 2.5×) was calculated and the weight in the retentate was then estimated by subtracting the permeate SP weight from the skim milk SP weight.

The rejection coefficient (σ) given by equation 1 was used to evaluate the overall MF membrane performance:

$$\sigma_{\text{CN or SP}} = 1 - \frac{C_P}{C_R}, \quad [1]$$

where C_P and C_R represent respectively permeate and retentate casein or SP concentrations. Ideally, σ_{CN} should be equal to 1 and σ_{SP} should be equal to 0 following MF of skim milk. The SP removal rate (kg/h per m²) was calculated by multiplying the permeate flux at a given VCF during MF by the SP concentration in the corresponding permeate sample.

Energy Consumption Measurement

Energy consumption was calculated from experimental measurements using a digital multimeter (modelr 3000FC, Fluke, Mississauga, ON, Canada) that recorded current voltage (U) and intensity (I) of the feed pumps and the retentate recirculation pump. Equation 2 was used to calculate the real power consumption (W_{applied}) of the 3-phase motors (Farooque et al., 2008):

$$W_{\text{applied}} = \sqrt{3} \times U \times I \times \eta, \quad [2]$$

where η is the pump efficiency. As all pumps had different efficiencies during operation, the η value was assumed to be 0.65, as reported by Adams and Barbano (2016). Equation 3 was applied to convert real power consumption to kilowatt-hours:

$$\text{kWh} = \frac{W_{\text{applied}} \times \text{Time (h)}}{1,000}. \quad [3]$$

Energy consumption data were used to compare the energy required to remove a kilogram of permeate (kWh/kg of permeate) and SP (kWh/kg of SP) in each experiment. In our system, the recirculation pump accounted for only 34% of all energy required, which is lower than expected, as the recirculation flow rate is much greater than the feed flow rate in a tubular configuration (Cheryan, 1998). This is due the simultaneous use of 2 feed pumps that were oversized so that the system could also be used for UF, nanofiltration, or reverse osmosis. In the dairy industry, a MF system for skim milk would obviously have more suitably sized pumps to reduce power consumption.

Membrane Fouling Characterization

Membrane fouling was characterized using the resistance-in-series model (Bae and Tak, 2005),

$$R_t = R_m + R_c + R_f, \quad [4]$$

where R_t is the total resistance (m⁻¹), R_m is the membrane resistance (m⁻¹), R_c is the reversible resistance (m⁻¹), and R_f is the irreversible resistance (m⁻¹). Reversible resistance is caused by the cake layer on the membrane surface, and irreversible resistance is due to pore plugging and irreversible adsorption of foulants onto the membrane (Bae and Tak, 2005). All types of resistances can be calculated using the following equations:

$$R_m = \frac{\Delta P_T}{\mu J_w}, \quad [5]$$

$$R_c = \frac{\Delta P_T}{\mu J_{AS}} - R_m, \quad [6]$$

and

$$R_f = \frac{\Delta P_T}{\mu J'_w} - R_m, \quad [7]$$

where ΔP_T is the TMP (Pa), μ the permeate viscosity (Pa·s), J_w is the clean water flux (m^3/m^2 per second), J'_w is the fouled water flux (m^3/m^2 per second), and J_{AS} is the permeate flux (m^3/m^2 per second) at the end of the MF procedure (Bae and Tak, 2005). Water has a μ value of 0.547 mPa·s (Weast et al., 1988), whereas MF permeate from skim milk has a μ value of 0.650 mPa·s (Adams et al., 2015). Although slight changes in permeate viscosity might occur during DF due to dilution with water, the value for MF permeate of skim milk was used.

For equivalent processing parameters (temperature, cross-flow velocity, TMP, feed composition, and so on), resistance is a good indicator of the type of fouling during filtration. For example, a high R_c -to- R_t ratio indicates that the drop in flux is caused mostly by caking, whereas a high R_f -to- R_t ratio indicates that the drop is caused mostly by pore plugging (Bae and Tak, 2005).

Statistical Analysis

Analyses were conducted using SAS University Edition (Version 3.4, SAS Institute Inc., Cary, NC). Significant differences between means were determined using the GLM procedure and Tukey tests with $P \leq 0.05$ as the criterion.

RESULTS

Determination of Optimal TMP for SP Removal

As shown in Figure 2, mean permeate flux increased linearly with TMP up to 179 kPa. The critical flux was reached apparently between 179 and 207 kPa, as the relationship between flux and TMP was no longer linear in this range, whereas the limiting flux was not reached at the pressure levels tested. At each pressure level, the permeate flux remained stable during the 45-min periods, meaning that no major membrane fouling occurred.

No significant differences ($P > 0.05$) were observed in the general composition of permeates and retentates

Figure 2. Permeate flux at different transmembrane pressures (TMP) during microfiltration of skim milk at 50°C. R1 = replicate 1; R2 = replicate 2. Flux was measured weighing the permeate collected over a period of 60 s.

recovered at different TMP (Table 1). However, the lowest SP rejection coefficient (0.359) was calculated for 124 kPa whereas the highest (0.472) was calculated for 207 kPa. This decrease in SP transmission suggests that the critical flux was indeed reached at TMP in the 179 to 207 kPa range.

On the basis of these results, 152 kPa was identified as the optimal TMP for obtaining a VCF of 3× in subsequent experiments. This pressure represented a compromise between high permeate flux and low SP rejection coefficient, in addition to being well below the critical flux. At a VCF of 3× the critical flux will decrease due to an increase in the feed concentration, but a TMP of 152 kPa should correspond to the sustainable flux, which is still acceptable for this process (Hurt et al., 2015a). The clean water flux was typically 395 kg/h per m^2 and the fouled water flux was 241 kg/h per m^2 . The water flux was always restored by cleaning.

SP Removal During Skim Milk Concentration and DF

A final VCF of 3× was chosen because this a common value sought in MF processing of skim milk using GP membranes (Zulewska et al., 2009; Zulewska and Barbano, 2014). The batch process used in our study allowed accurate determination of flux and composition during skim milk concentration and DF at an average TMP of 152 kPa. As shown in Figure 3, permeate flux was highest at a VCF of 1× and decreased to approximately 80% of this value a VCF of 2×. At a VCF of 3×, permeate flux was approximately 53% of the initial flux.

Changes in permeate and retentate compositions during the concentration of skim milk are reported in Table 2. Significant differences were observed among retentates as a result of casein concentration as a function of VCF. Permeate contained significantly more SP

Table 1. Mean composition¹ (% by weight) of skim milk and microfiltration permeate and retentate produced at different transmembrane pressure (TMP)^{2,3}

TMP (kPa)	Sample	TN (%)	NCN (%)	NPN (%)	TP (%)	CN (%)	SP (%)	CN%TP	Solids (%)	SP removal rate (kg/h per m ²)
—	Skim milk	3.20	0.67	0.16	3.05	2.53	0.51	83.20	8.52	—
124	Retentate	3.40	0.69	0.17	3.24	2.71	0.53	83.67	8.94	0.40
	Permeate	0.55	0.50	0.16	0.39	0.05	0.34	13.02	5.83	
152	Retentate	3.47	0.71	0.16	3.31	2.76	0.55	83.36	8.96	0.50
	Permeate	0.54	0.49	0.16	0.38	0.05	0.33	13.84	5.84	
179	Retentate	3.44	0.69	0.16	3.27	2.75	0.52	83.95	8.86	0.58
	Permeate	0.53	0.49	0.16	0.37	0.05	0.32	12.69	5.82	
207	Retentate	3.50	0.68	0.18	3.32	2.82	0.51	84.72	9.03	0.52
	Permeate	0.49	0.43	0.16	0.33	0.06	0.27	18.76	5.77	

¹Based on duplicate experiments.²TN = total nitrogen $\times$ 6.38; NCN = noncasein nitrogen $\times$ 6.38; NPN = nonprotein nitrogen $\times$ 6.38; TP = true protein (TN – NPN); CN = casein (TN – NCN); SP = serum proteins (TP – CN); CN%TP = casein as percentage of true protein.³Values in columns were not different (Tukey, $P > 0.05$). Permeates are compared with permeates and retentates are compared with retentates.

at a VCF of 3 $\times$ than at 1.5 $\times$. However, the retentates also contained more SP, meaning that SP rejection coefficient increased overall as the VCF increased to 0.441, 0.462, 0.554, and 0.559 at VCF of 1.5, 2, 2.5, and 3 $\times$, respectively. The increase was considerable between VCF of 2 and 2.5 $\times$. As shown in Table 2, the SP removal rate tended to decrease as VCF increased. As a result of differential transmission of SP and casein, the casein as a percent of TP value increased from 82.87% in skim milk to 89.59% in the retentate at a VCF of 3 $\times$. However, no significant difference was noted in casein as a percent of TP between VCF of 2 and 3 $\times$, which may indicate that retentate cannot be further purified without DF. The permeate compositions reported in Table 2 also show that some casein (0.05–0.06%) passes through no matter the VCF. The clean water flux was typically 390 kg/h per m² and the fouled water flux was 267 kg/h per m². Again, cleaning always restored the water flux.

Changes in permeation fluxes during DF are illustrated in Figure 4. The flux declined during the initial concentration, in a manner similar to that noted in the concentration experiment. At VCF of 3 $\times$, the mean permeate flux was approximately 61% of the initial level. After the first addition of 173 kg of tap water (DF 1), the flux was restored to 94% of the initial flux. The flux was about 78% of the initial level when the VCF reached 3 $\times$. After the second addition of 173 kg of tap water (DF 2), the flux was restored to 97% of the initial water flux. It decreased slightly during the last concentration to 94% of the initial water flux at VCF of 3 $\times$.

Table 3 shows that TN, NCN, NPN, TP, SP, and solids decreased ($P \leq 0.05$) in the permeate between the different stages of processing, due mostly to dilution by the DF water. For the retentate, significant differences were observed for NPN and solids between the different

stages of processing. This is due to NPN and other solids (mostly lactose) consisting of molecules too small to be rejected by the membrane. The casein as a percent of TP value was expected to increase between DF 1 and 2, as reported in the literature (Zulewska and Barbano, 2014), but remained essentially unchanged (Table 3). The clean water flux was typically 365 kg/h per m² and the fouled water flux was 272 kg/h per m².

Energy Consumption Measurement

In the full recycle experiment (Table 4), it was found that as the TMP increased, the kilowatt-hours per kilogram of permeate decreased significantly, except between TMP of 179 and 207 kPa ($P > 0.05$). However, no significant effect of TMP was noted on kilowatt-hours per kilogram of SP removed. In addition, the energy required tended to decrease as the TMP increased. This is because the permeate SP content remained rela-

Figure 3. Permeate flux at different volumetric concentration factors during microfiltration of skim milk at 50°C. R1 = replicate 1; R2 = replicate 2. Flux was measured by weighing the permeate collected over a period of 60 s.

Table 2. Mean composition¹ (% by weight) of skim milk and microfiltration permeate and retentate produced at different volumetric concentration factor (VCF)²

VCF	Sample	TN (%)	NCN (%)	NPN (%)	TP (%)	CN (%)	SP (%)	CN%TP	Solids (%)	SP removal rate (kg/h.m ²)
—	Skim milk	3.29	0.70	0.16	3.12	2.59	0.54	82.87	9.05	—
1.5×	Retentate	4.83 ^a	0.82 ^a	0.16 ^a	4.67 ^a	4.00 ^a	0.66 ^a	85.76 ^a	10.82 ^a	0.50 ^a
	Permeate	0.59 ^a	0.54 ^a	0.16 ^a	0.43 ^a	0.06 ^a	0.37 ^a	13.50 ^a	6.11 ^a	
2×	Retentate	6.24 ^b	0.87 ^a	0.16 ^a	6.08 ^b	5.37 ^b	0.71 ^a	88.26 ^b	12.41 ^b	0.45 ^a
	Permeate	0.61 ^a	0.55 ^{ab}	0.17 ^a	0.44 ^{ab}	0.06 ^a	0.38 ^{ab}	12.76 ^a	6.15 ^a	
2.5×	Retentate	7.60 ^c	1.03 ^b	0.16 ^a	7.44 ^c	6.56 ^c	0.88 ^b	88.22 ^b	14.02 ^c	0.36 ^b
	Permeate	0.61 ^a	0.56 ^b	0.17 ^a	0.44 ^{ab}	0.05 ^a	0.39 ^{ab}	12.02 ^a	6.18 ^a	
3×	Retentate	9.02 ^d	1.08 ^b	0.16 ^a	8.86 ^d	7.94 ^d	0.92 ^b	89.59 ^b	15.31 ^d	0.32 ^b
	Permeate	0.62 ^a	0.57 ^b	0.17 ^a	0.46 ^b	0.05 ^a	0.41 ^b	11.15 ^a	6.18 ^a	

^{a-d}Values in the same column without a common superscript are significantly different (Tukey, $P \leq 0.05$). Permeates are compared with permeates and retentates are compared with retentates.

¹Based on duplicate experiments.

²TN = total nitrogen $\times 6.38$; NCN = noncasein nitrogen $\times 6.38$; NPN = nonprotein nitrogen $\times 6.38$; TP = true protein (TN – NPN); CN = casein (TN – NCN); SP = serum proteins (TP – CN); CN%TP = casein as percentage of true protein.

tively stable as TMP changed, whereas permeate flux increased, thus shortening the time required to obtain 1 kg of protein.

In the concentration experiment, significant differences were observed in kilowatt-hours per kilogram of permeate and per kilogram of SP transmitted as a function of VCF at constant TMP. Table 4 shows that energy consumption increased constantly until the VCF reached 3 $\times$. However, no significant difference was observed between VCF of 1 up to 2.5 $\times$. Although the MF was conducted in batch mode, the experiment can be compared with continuous MF because both are presented as processing steps. Permeate fluxes obtained at different VCF (Figure 3) can be compared using the corresponding energy consumption values in Table 4.

Energy consumption calculated in the concentration/DF experiment reveals that differences in kilowatt-hours per kilogram of permeate between the initial concentration process and DF 2 are relatively small

but significant, due to an increase in permeate flux between processing steps. However, the differences in kilowatt-hours per kilogram of SP transmitted are more substantial and show that the first and second DF steps require respectively up to 2 and 5 times more energy than initial concentration. This large increase in energy consumption is due mainly to the lower SP content of DF permeates because of dilution with water. Whereas the energy consumption findings may not be applicable to every MF system, tendencies similar to those described in Table 4 are likely to be observed under other circumstances and are therefore relevant.

DISCUSSION

Flux Performance of GP Membrane

The fluxes reported in the present study are in general agreement with published data. The average flux at a VCF of 3 $\times$ differed from one experiment to the next: 79.1 kg/h.m² in the concentration experiment and 90.0 kg/h per m² in the concentration/DF experiment (before DF). These differences are due likely to variation in the TS contents of the batches of milk used and the VCF actually achieved at the end of filtration, as VCF is more difficult to estimate in batch mode. The fluxes were nevertheless slightly higher than those obtained in similar studies—72.5 kg/h per m² by Zulewska and Barbano (2014) and 71.8 kg/h per m² by Zulewska et al. (2009)—whereas the overall pressures maintained (Rpi, Rpo, and Ppo) were lower. The same tendency was noted during DF, the flux reaching, on average, 115.4 kg/h per m² after DF 1 and 139.3 kg/h per m² after DF 2, compared with 84.5 and 92.7 kg/h per m² in a similar experiment conducted by Zulewska and Barbano (2014).

Figure 4. Permeate flux during initial concentration and subsequent diafiltration (DF) of skim milk at 50°C R1 = replicate 1; R2 = replicate 2; VCF = volumetric concentration factor. Flux was measured by weighing the permeate collected over a period of 60 s.

Table 3. Mean composition¹ (% by weight) of skim milk and microfiltration permeate and retentate produced at different stages of concentration and diafiltration (DF)²

Stage of processing	Sample	TN (%)	NCN (%)	NPN (%)	TP (%)	CN (%)	SP (%)	CN%TP	Solids (%)	SP removal rate (kg/h.m ²)
—	Skim milk	2.96	0.65	0.16	2.80	2.31	0.49	82.58	8.52	—
After initial concentration	Retentate	7.79 ^a	1.00 ^a	0.15 ^a	7.64 ^a	6.79 ^a	0.85 ^a	88.91 ^a	13.77 ^a	0.44 ^a
	Permeate	0.56 ^a	0.51 ^a	0.16 ^a	0.40 ^a	0.06 ^a	0.35 ^a	14.45 ^a	5.79 ^a	
After DF 1	Retentate	7.45 ^a	0.71 ^b	0.06 ^b	7.40 ^a	6.74 ^a	0.65 ^b	91.20 ^b	9.90 ^b	0.18 ^b
	Permeate	0.22 ^b	0.18 ^b	0.05 ^b	0.17 ^b	0.04 ^a	0.13 ^b	21.94 ^a	1.92 ^b	
After DF 2	Retentate	7.12 ^a	0.63 ^b	0.03 ^c	7.09 ^a	6.49 ^a	0.60 ^b	91.57 ^b	8.92 ^c	0.09 ^c
	Permeate	0.11 ^c	0.08 ^c	0.02 ^c	0.09 ^c	0.03 ^a	0.06 ^c	33.79 ^a	0.68 ^c	

^{a-c}Values in the same column without a common superscript are significantly different (Tukey, $P \leq 0.05$). Permeates are compared with permeates and retentates are compared with retentates.

¹Based on duplicate experiments.

²TN = total nitrogen $\times 6.38$; NCN = noncasein nitrogen $\times 6.38$; NPN = nonprotein nitrogen $\times 6.38$; TP = true protein (TN – NPN); CN = casein (TN – NCN); SP = serum proteins (TP – CN); CN%TP = casein as percentage of true protein.

A decrease in SP removal rate (Table 2) during skim milk concentration experiments indicated build-up of a fouling layer as the VCF was increased. Previous study showed that, at a VCF of 3 $\times$, GP membrane achieved a SP removal rate of 0.37 kg/h per m² (Zulewska and Barbano, 2014). The results obtained in the present study are therefore consistent with those reported in the literature. The SP removal rate dropped significantly during DF steps (Table 3). Even though adding water increased the permeate flux, dilution was occurring at the same time, thus decreasing the overall efficiency of the process. The weight of permeate required to remove 1 kg of SP increased as tap water was added to skim milk. This value was 290 kg at VCF of 3 $\times$ after the initial concentration, 770 kg after DF 1, and 1,628 kg after DF 2. In other words, DF increases considerably the volume of the stream from which the SP must be recovered.

Membrane Fouling

Clean water flux was slightly lower in the concentration/DF experiment than in the other experiments, which explains the significantly higher ($P \leq 0.05$) calculated membrane resistance (R_m) value (Figure 5). However, the water flux was still within the acceptable range (within 10% of the initial water flux). At the end of the concentration experiment, the R_c -to- R_t ratio (0.593) was higher than at the end of the concentration/DF experiment (0.300), meaning that the fouling layer was thicker. The DF water decreased the TS content of the retentate and, hence, the thickness of the fouling layer, which increased the permeate flux and reduced the R_c -to- R_t ratio measured at the end of the experiment. No significant difference in irreversible resistance was observed between experiments. This may indicate that most of the pore plugging appeared during the

Table 4. Power consumption during microfiltration of skim milk at 50°C under different transmembrane pressure (TMP), volumetric concentration factors (VCF), and diafiltration (DF) conditions

Item	Level	Feed + recirculation pumps ¹	
		kWh/kg of permeate	kWh/kg of SP transmitted
TMP	124 kPa	0.094 ^a	28.29 ^a
	152 kPa	0.073 ^b	22.45 ^a
	179 kPa	0.062 ^c	19.27 ^a
	207 kPa	0.057 ^c	21.54 ^a
VCF reached (at TMP = 152 kPa)	1–1.5 $\times$	0.093 ^a	24.92 ^a
	1.5–2 $\times$	0.099 ^a	25.73 ^a
	2–2.5 $\times$	0.113 ^{ab}	28.99 ^{ab}
	2.5–3 $\times$	0.143 ^b	34.98 ^b
Concentration/DF to VCF = 3 $\times$ (TMP = 152 kPa)	Concentration	0.089 ^a	25.83 ^a
	DF #1	0.083 ^{ab}	63.99 ^b
	DF #2	0.077 ^b	124.85 ^c

^{a-c}Means in the same column without a common superscript are significantly different (Tukey, $P \leq 0.05$). Comparisons were made with results from the same experiment only.

¹The recirculation pump accounted for 34% of all energy required.

initial concentration to VCF of 3 $\times$ and could not be reversed by adding tap water.

SP and Casein Transmission

As the goal of this MF process is to retain all casein in the retentate, it is important to measure the quantity that crossed the membrane. Those casein are considered lost because they cannot be used for cheese-making, thus decreasing the profitability of the process. The transmission of casein through GP membrane has been found higher than through spiral-wound or UTP membranes (Zulewska et al., 2009). As shown in Figure 6a, approximately 1.4% of the total casein crossed the membrane at a VCF of 3 $\times$. Diafiltration increased this loss to 2.8 and 3.8% (DF 1 and 2, respectively; Figure 6b). It has been found previously that DF allows more casein to pass through the membrane than simple concentration (Zulewska and Barbano, 2014). During our concentration/DF experiment, the permeate casein concentration was relatively constant from one stage

to the next whereas the SP concentration decreased, which indicates that the weight of SP removed by 1 kg of casein loss decreased. At a VCF of 3 $\times$, this ratio was 5.9, 3.6, and 2.0 kg, respectively, after initial concentration, DF 1, and DF 2, indicating increased contamination of permeate by casein under conditions of DF.

In terms of the percentage of SP removed at the end of processing (Figure 6), results were better in previous studies, such as 56.0% in Zulewska and Barbano (2014) and 61.0% in Zulewska et al. (2009), whether compared with the concentration (47.6%) or the concentration/DF (47.0% before DF) experiments. This might be due to a higher SP rejection coefficient in our experiment, which could result from higher permeation fluxes (closer to the limiting flux) or to a different start-up procedure. Again, SP removal from skim milk was 64.7% after DF 1 and 73.2% after DF 2, compared with 82.6 and 96.5% obtained by Zulewska and Barbano (2014) after similar DF steps. Although the differences appear considerable, this mass balance calculation might not be the best way to evaluate the efficiency of the process. These calculations overestimate the quantity of SP transmitted because they do not take into consideration the amount remaining in the retentate, thus reducing the accuracy of the mass balance.

Process Efficiency

Process efficiency was evaluated in terms of SP removal rate, energy consumption, total SP removed from skim milk, weight of permeate required to remove 1 kg of SP, weight of SP removed by 1 kg of casein loss in the permeate, SP rejection coefficient, casein as a percent of TP value of the retentate, membrane resistance, and permeate flux. None of these parameters indicated clearly what the optimal VCF was or whether or not DF improved process efficiency. The 3 most important parameters from a dairy processing perspective were then selected for the comparison of the experimental conditions: total energy consumption, cumulative loss of casein, and SP remaining in the retentate. All values were converted to percentages of the maximum value to make a 3-axis figure in which the lowest value represents the best option. Figure 7a shows clearly that VCF of 1.5 $\times$ has a major advantage in terms of 2 of the 3 parameters, whereas VCF of 3 $\times$ is the worst for 2 parameters but best in terms of SP remaining in the retentate. Figure 7b shows that simple concentration (to VCF of 3 $\times$) is the best in terms of total energy consumption, but leaves more SP in the retentate than does either DF. For example, the second DF step required 45% more energy and resulted in 35% more casein loss in the permeate, whereas only 13%

Figure 5. Reversible (R_c), irreversible (R_f), and membrane resistance (R_m) after initial concentration of skim milk to volumetric concentration factor (VCF) = 3 $\times$ (A) and reconcentration to VCF = 3 $\times$ after 2 diafiltrations (B). Means in the same type of resistance without a common letter (a,b) are significantly different (Tukey, $P \leq 0.05$).

Figure 6. Mass balance of casein (CN) and serum proteins (SP) at (A) different volumetric concentration factors (VCF) and (B) at VCF = 3× after initial concentration then first and second diafiltration (DF).

more SP was removed from the retentate compared with the first DF step.

It is not possible to compare Figures 7a and 7b because the units are expressed in relative percentage, but it is reasonable to expect a lower VCF to require less total energy and result in a smaller total loss of casein but a higher retentate SP content compared with DF. Dairy processors of course might not all attach the same value to each of these 3 parameters. Depending on economic conditions, some might value transmitted SP more whereas some might consider casein loss or total energy cost to be more important. In other words, these figures can be used to assess the overall process efficiency, but do not give a specific answer, as each dairy-processing facility has its own economic reality.

In all cases, the total amount of permeate generated must be taken into consideration. For simple concentration at VCF of 3×, 2 kg are generated for each kilogram of retentate, whereas 4 and 6 kg of permeate are generated for each kilogram of retentate after 1 and 2 DF, respectively. A dairy processor must be able to reduce the environmental impact of this waste stream efficiently. For example, cheese whey has a biochemical

oxygen demand of 40 to 60 g/L and a chemical oxygen demand of 50 to 80 g/L, lactose being responsible for 90% of both values (Kim et al., 2006). Although the composition of MF permeate differs from that of cheese whey, its lactose content is similar. Means of valorization of the MF permeate were not evaluated in our study, but it is clear that more permeate equals more processing costs because it mostly contains water and that lactose and SP must be further purified (evaporation, filtration, spray-drying, crystallization, and so on) to gain value.

Efficient Process Proposal

As stated by Zulewska and Barbano (2014), operating at higher flux improves the economic feasibility of the MF process by decreasing the membrane surface area requirement. Membrane surface area represents a huge capital cost, particularly for GP membrane (approximately \$3,100/m², excluding module). A larger membrane surface area also means higher operating costs (e.g., for cleaning, maintenance, and pumping) and greater losses of milk due to dilution in the filtra-

Figure 7. Relative percentages of cumulative kilowatt-hours used, casein (CN) loss in the permeate, and serum proteins (SP) in the retentate (A) at different volumetric concentration factors (VCF) and (B) at VCF = 3 $\times$ after initial concentration then first and second diafiltration (DF).

tion system dead volume at the beginning and end of production (Skrzypek and Burger, 2010).

As cheese yield is directly proportional to the casein content of the cheese milk (Neocleous et al., 2002a), casein-enriched MF retentate can be added in different proportions to standardize cheese milk. Table 5 shows that, for a given final casein concentration of cheese milk, the proportion of MF retentate required decreases as the VCF of the retentate increases whereas the permeate and SP obtained change very little. The membrane surface area required increases with VCF of the MF retentate due to decreasing permeate flux. As the initial whole milk skimmed for the production of MF retentate does not provide enough fat for cheesemaking, additional cream is required for standardization of the cheese milk. The loss of casein in the permeate is expected to be similar at each VCF, as casein content is stable (Table 2), but energy consumption may increase at a higher VCF because of the lower permeate flux and the larger size of the MF system. The required VCF of

MF retentate increases with the required casein concentration of the cheese milk, resulting in increased permeate and SP production. Regardless of the cheese milk standardization scenario being examined, many factors must be considered to evaluate the economic feasibility of the process, including the amount of permeate produced, the current value of SP, the costs associated with membrane surface area, and the benefit associated with the increased productivity of the cheesemaking process. The membrane surface area requirements in Table 5 are based on permeate fluxes in Figure 3, and, as mentioned, permeate flux could be lowered to be closer to the critical flux to get a lower SP rejection coefficient. This would probably increase the surface area required, but at the same time more SP would be obtained for the same final casein concentration in the cheese milk.

Table 5 suggests that the best way to reduce the membrane surface area requirement is to improve flux by using a lower VCF, which improves the pro-

Table 5. Proportions of whole milk, cream, and different microfiltration retentates required to standardize 1 million kg of cheese milk at different casein concentrations in an 18-h run

Casein concentration of cheese milk ¹ (g/L)	VCF ² of MF retentate	Input (kg)			Whole milk supply required	Additional cream required	Output (kg)		Membrane surface area required (m ²)
		Whole milk ³	Cream ³	MF retentate ⁵			MF permeate	SP in the permeate	
32.3	1.5×	462,941	76,300	460,759	1,221,786	8,593	230,379	856	94.9
	2×	730,246	51,841	217,913	1,208,766	9,146	217,913	838	102.6
	2.5×	806,015	44,908	149,076	1,215,216	8,398	223,615	873	141.8
	3×	849,741	40,908	109,351	1,209,933	8,770	218,703	890	153.6
37.7	2×	471,676	95,277	433,047	1,422,615	10,431	433,047	1,664	204.0
	2.5×	622,249	81,500	296,252	1,435,433	8,945	444,378	1,735	281.9
	3×	709,143	73,549	217,309	1,424,933	9,684	434,617	1,768	305.2
43.0	2×	213,107	138,712	648,181	1,636,465	11,716	648,181	2,491	305.3
	2.5×	438,482	118,091	443,427	1,655,650	9,491	665,141	2,597	421.9
	3×	568,544	106,190	325,266	1,639,934	10,597	650,532	2,647	456.8
48.4	2.5×	254,715	154,682	590,603	1,875,867	10,038	885,905	3,458	561.9
	3×	427,946	138,831	433,223	1,854,935	11,511	866,446	3,525	608.4

¹CN-to-fat ratio: 0.68, as recommended by Neocleous et al. (2002a).²VCF = volumetric concentration factor.³Whole milk: 3.7% fat, 2.7% casein.⁴Cream: 40.0% fat, 1.8% casein.⁵Casein content from Table 2, no fat.

cess efficiency. In fact, studies have shown that the MF retentate can be used for cheese production, and, although the results are variable, a lower casein concentration factor offers better results (Neocleous et al., 2002b; Schreier et al., 2010). In their evaluation of the economic feasibility of using MF-processed skim milk for cheesemaking, Papadatos et al. (2003) concluded that increasing the VCF beyond 2× provides no additional benefit because of the higher processing cost. Nevertheless, those authors showed that the use of MF retentate was more cost-efficient than conventional cheesemaking and other milk fortification methods, such as the use of condensed milk or nonfat dry milk. However, their study focused primarily on the addition of MF retentate with a VCF of 2 or 3× to raw milk and cream to produce the equivalent of a retentate with a VCF of 1.25×. The conclusions of Papadatos et al. (2003) could be strengthened now that the superiority of GP membranes over UTP systems has been proven for MF of skim milk. Considering those factors, dairy processors should view MF as a way of adding value to the conventional cheesemaking process by producing a SP-enriched permeate and a casein-enriched retentate which could replace the use of UF to increase protein concentration of cheese milk. Compared with UF, MF allows higher permeate flux and requires lower operating pressures.

If the desired fraction is MCC, DF should be used to obtain a retentate with as little SP as possible. Although casein products such as MCC are being replaced progressively by milk protein concentrates or isolates (Lagrange et al., 2015), the literature on

their production is abundant. Zulewska and Barbano (2014) estimated that the use of GP membrane allows reduction of the total surface area needed to process 1 million kg of skim milk (over an 18-h run) to 1,208 m², compared with 2,051 m² for a UTP system. They estimated that this process would allow removal of more than 95% of the SP from the milk, but that 2 DF steps would be needed, meaning that, overall, 1 million kg of retentate and 2 million kg of permeate would be generated (Zulewska and Barbano, 2014). Although such a process is theoretically feasible, it would be less efficient than the standardization of cheese milk with MF retentate. Also, dairy processor should have the equipment needed to manage all those effluents.

CONCLUSIONS

This study presents an analysis that will help dairy processors to identify the key parameters for optimizing their investment in a GP membrane system. Whereas achieving a higher VCF or using DF allows the separation of more SP from skim milk, these approaches do not necessarily result in a more efficient process. In fact, a lower VCF allows higher permeate flux and a lower SP rejection coefficient, while requiring less energy and reducing casein loss. Diafiltration is a viable option if the dairy processor wants to produce MCC, as it allows separation of almost all of the SP from skim milk. Milk used for cheesemaking should be standardized with MF retentate made at the lowest possible VCF. Dairy processors should use skim milk MF retentate to increase cheese production capacity, while

generating permeate with added value compared with cheese whey. Although the results of this study show some tendencies regarding the efficiency of processing skim milk using GP MF membrane, the economic feasibility of the process remains the main factor that will determine whether or not this technology should be used at a large scale.

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