The effect of linear velocity and flux on performance of ceramic graded permeability membranes when processing skim milk at 50°C¹

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ABSTRACT

Raw milk (about 500 kg) was cold (4°C) separated and then the skim milk was pasteurized at 72°C and a holding time of 16 s. The milk was cooled to 4°C and stored at ≤4°C until processing. The skim milk was microfiltered using a pilot-scale ceramic graded permeability (GP) microfilter system equipped with 0.1-μm nominal pore diameter ceramic Membralox membranes. First, about 155 kg of pasteurized skim milk was flushed through the system to push the water out of the system. Then, additional pasteurized skim milk (about 320 kg) was microfiltered (stage 1) in a continuous feed-and-bleed 3× process using the same membranes. The retentate from stage 1 was diluted with pasteurized reverse osmosis water in a 1:2 ratio and microfiltered (stage 2) with a GP system. This was repeated 3 times, with total of 3 stages in the process (stage 1 = microfiltration; stages 2 and 3 = diafiltration). The results from first 3 stages of the experiment were compared with previous data when processing skim milk at 50°C using ceramic uniform transmembrane pressure (UTP) membranes. Microfiltration of skim milk using ceramic UTP and GP membranes resulted in similar final retentate in terms of serum proteins (SP) removed. The SP removal rate (expressed by kilogram of SP removed per meter-squared of membrane area) was higher for GP membranes for each stage compared with UTP membranes. A higher passage of SP and SP removal rate for GP than UTP membranes was achieved by using a higher cross-flow velocity when processing skim milk. Increasing flux in subsequent stages did not affect membrane permeability and fouling. We operated under conditions that produced partial membrane fouling, due to using a flux that was less than limiting

flux but higher than critical flux. Because the critical flux is a function of the cross-flow velocity, the difference in critical flux between UTP and GP membranes resulted only from operating under different cross-flow velocities (6.6 vs 7.12 for UTP and GP membranes, respectively). Conditions that allow microfiltration operation at higher flux will reduce the membrane surface area required to process the same amount of milk in the same length of time. Less membrane surface area reduces investment costs and uses less energy, water, and chemicals to clean the microfiltration system.

Key words: microfiltration, linear velocity, flux, serum protein removal

INTRODUCTION

One of the main limitations in application of microfiltration (MF) membranes in the dairy industry is fouling. Fouling phenomena in microfiltration and its effect on flux and protein separation was the subject of several reviews (Marshall et al., 1993; Brans et al., 2004). Direct evidence of fouling during the filtration process is flux decline. Several phenomena reduce flux, namely, concentration polarization, protein deposition, and consolidation of the fouling material. Different strategies have been developed to reduce the effect of fouling. Wakeman and Williams (2002) indicated that the most popular were feed pretreatment, choice of membrane material, flow manipulation, the use of high shear, and gas sparging. Some methods for controlling fouling are effective, whereas some still need improvements. Depending on the specific commercial application, a different approach for fouling control may be required (Brans et al., 2004).

Several module modifications were designed to improve the MF performance. The uniform transmembrane pressure (UTP) approach, developed by Sandblom (1978), results in low transmembrane pressure (TMP) and uniform flux along the length of the membrane. The UTP concept overcomes most of the heterogeneous fouling of the MF membrane, which results from the high cross-flow velocities required for obtaining both high permeation fluxes and accurate

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selectivity. However, this approach has some disadvantages, such as high investment and operating costs as a result of recirculation pumping of permeate to produce co-current permeate flow. Nevertheless, in MF of skim milk to separate CN micelles from serum proteins (**SP**), UTP has been used commercially (Brans et al., 2004). Alternative methods to obtain uniform flux with lower operational costs include graded permeability (Garcera and Toujas, 2002) and Isoflux (Grangeon et al., 2002) ceramic membranes. These membranes have a decreasing permeability built into either the external surface of the support structure (GP membranes) or selective layer (Isoflux) on the inside of the flow channels. The comparative studies (Zulewska et al., 2009; Hurt et al., 2010; Adams and Barbano, 2013) carried out in our laboratory showed differences in membrane selectivity and process yield when processing skim milk with different ceramic modules. In general, UTP system showed best SP passage, followed by GP and Isoflux membranes.

It is typical to operate ceramic MF membranes at very high cross-flow velocities (Altmann and Ripperger, 1997). Wall shear stress (τ_w), which is directly proportional to cross-flow velocity (Samuelsson et al., 1997), creates shear turbulent flow that minimizes fouling and produces a separation of CN and SP with ceramic membranes. Increasing cross-flow velocity may result in higher passage of SP and SP removal rate (expressed by kilograms of SP removed per meter-squared of membrane area). The separation process is very dependent on the forces acting on a single particle and is, to a large extent, influenced by the balance between the lift force and the drag force acting on the particles in the feed stream (Altmann and Ripperger, 1997). If the inertial lift velocity is lower than permeate velocity, then the particles tend to deposit on the membrane surface (Belfort et al., 1994). Under fast laminar flow conditions, a cubic effect of the particle size and squaring of tangential shear rate on the increase in inertial lift velocity exists; thus, in case of large particles and high flow rate, the effect is more prominent (Belfort et al., 1994). In general, flux and wall shear stress are 2 major parameters of the separation process that generate the lift and drag forces. Permeation flux rules the transport of mass to the membrane (convection) and tangential flow velocity, through the wall shear stress, drives transport particles back toward the bulk (erosion, hydrodynamic diffusion, or lateral migration of particles due to an inertial force; Le Berre and Daufin, 1996). The critical flux is defined as the flux above which irreversible fouling occurs (Bacchin, 2004). Flux determines the yield of the separation process (when processing at values less than the critical flux), not the permeability and selectivity (Le Berre and Daufin, 1996). Wall shear

stress has both a quantitative and qualitative effect. Higher shear stress causes a thinner deposit layer on the membrane surface and higher permeation flux, however, an excessive increase in shear stress may lead to lower SP passage (Le Berre and Daufin, 1996). Le Berre and Daufin (1996) suggested that membrane filtration performance, especially the physical conditions at the interface membrane or feed, greatly depends on the ratio of flux to tangential shear stress. The ratio defines the balance of particle deposition rate (relating to flux) and particle removal rate (relating to shear stress; Gesan-Guizieu et al., 2000). To achieve satisfactory MF performance (i.e., minimum fouling at high protein transmission), it is recommended to operate under conditions in which the ratio of flux to wall shear stress is lower than a critical value (Grandison et al., 2000). The objective of our research was to determine if increased linear velocity and retentate and permeate removal rates will increase the productivity (i.e., SP removal rate) without increasing fouling when processing skim milk with 0.1- μ m Membralox GP membranes.

MATERIALS AND METHODS

Experimental Design and Statistical Analysis

Raw milk (about 500 kg) was cold (4°C) separated and then pasteurized at 72°C and a holding time of 16 s. The milk was cooled to 4°C and stored at $\leq 4^\circ\text{C}$ until processing. The skim milk was MF using pilot-scale ceramic graded permeability (**GP**) MF system equipped with 0.1- μ m nominal pore diameter ceramic Membralox membranes (model EP1940GL0.1 μ AGP1020, Alumina, Pall Corp., East Hills, NY). First, about 155 kg of skim milk was flushed through the system to remove the water from the system. Then, additional pasteurized skim milk (about 320 kg) was MF (stage 1) in a continuous feed-and-bleed 3 $\times$ process using the same membranes. The retentate from stage 1 was diluted with pasteurized reverse osmosis (**RO**) water in a 1:2 ratio and MF (stage 2) with the GP system. The complete process had a total 3 stages (stage 1 = MF; stages 2 and 3 = diafiltration). The results from first 3 stages of the GP process in the present study were compared with the data reported by Hurt et al. (2010) when processing skim milk at 50°C using a 3-stage ceramic UTP membrane process.

To determine if significant differences existed between GP and UTP membranes, the data were analyzed by ANOVA using the Proc GLM procedures of SAS (SAS version 8.02, SAS Institute Inc., Cary, NC) and the GLM model was dependent variable = treatment (GP or UTP) + error. The GLM model dependent variable = stage of processing + replicate + error was used

to determine if a significant difference in SP removal from skim milk due to stage of processing existed. A similar GLM model was used to determine if flux at the inlet end of the membrane, transmembrane pressure inlet (**TMP_{in}**), outlet end of the membrane, transmembrane pressure outlet (**TMP_{out}**), and difference in pressure (ΔP) changed with the time of processing run for all stages. Time was treated as a continuous variable in the split-plot ANOVA design. Distortion of the ANOVA by multicollinearity in the model was minimized by centering the time of running using a mathematical transformation (Glantz and Slinker, 2001). Time was transformed as time transformed = running time - [(last time - first time)/2]. This transformation made the data set orthogonal with respect to time. The model was dependent variable (flux, **TMP_{in}**, **TMP_{out}**, or ΔP) = stage + replicate + stage $\times$ replicate + time + time $\times$ replicate + time $\times$ stage + error, with stage and replicate as category variables and time as a continuous variable.

Production of 95% SP Reduced Micellar CN Concentrate

A pilot-scale GP MF system (Tetra Alcross MFS-7, TetraPak Filtration Systems, Aarhus, Denmark) equipped with 0.1- μ m nominal pore diameter ceramic Membralox membranes (surface area: 1.7 m²) was used. The membranes in a tubular single stainless steel module consisted of 7 ceramic tubes, 19 channels each with a 4-mm channel diameter. No beads were present on the permeate side of the GP membrane. The GP MF system consisted of a feed pump (type LKH 10/110 SSS 1.75 kW) and a retentate recirculation pump (type LKH 20/125 SSS 6.3 kW), both from Alfa Laval (Kansas City, MO). The membranes were 1.02 m long and were mounted vertically in the MF system with inlet at the top and outlet at the bottom of the vertically mounted membrane. Permeate was removed at the top of the vertical membrane module with back pressure applied. The same cleaning procedure as described by Zulewska et al. (2009) was applied before processing.

Because the membrane was mounted vertically, the inlet and outlet gauge pressures had to be corrected for the difference because of the weight of the vertical column of liquid. The correction was measured with 50°C RO water in the system and only the feed pump turned on and the retentate and permeate outlet valves closed. The retentate pressure inlet (**R_{pi}**), the permeate pressure inlet (**P_{pi}**; at the retentate inlet end of the membrane), and the retentate pressure outlet (**R_{po}**) were measured under these conditions. The permeate pressure outlet (**P_{po}**; at the retentate outlet end of the membrane) was not measured directly and was

calculated by adding 13.79 kPa to the permeate pressure inlet, which was the correction for the elevation difference (102 cm) and weight of the column of liquid between the inlet and outlet end of the membrane. The correction factor was calculated for each gauge pressure as P_{po} minus R_{pi} for the R_{pi} gauge pressure correction; P_{po} minus R_{po} for the R_{po} gauge pressure correction; P_{po} minus P_{pi} for the P_{pi} gauge pressure correction; and the P_{po} gauge pressure correction was zero.

After gauge correction, the retentate recirculation pump was turned on and the retentate and permeate removal rates were set to 1,020 and 2,040 g per 60 s, which corresponded to approximately 60 and 120 L/h on the flowmeters, respectively, to achieve 3 $\times$ concentration factor (**CF**). The cross-flow velocity was adjusted to 7.12 m/s and the feed for the system was switched from water to skim milk.

Flushing Water from the System. First, about 155 kg of pasteurized skim milk at 50°C was MF using the GP membranes to flush the water from the system at start-up of the first stage of the process. With the retentate and permeate removal rates set at 60 and 120 L/h, the average flushing time was 45 min. The retentate and permeate removal rates were set to CF 3.00 $\pm$ 0.05 as measured based on the weights to the nearest gram of the retentate and permeate collected in 60 s (SB32000, Mettler Toledo, Columbus, OH). If necessary, the retentate and permeate removal rates were adjusted to maintain a 3 $\times$ CF. Samples of permeate and retentate were taken every 10 min during this initial stage and protein, fat, and lactose concentrations were determined using an midinfrared spectrophotometer (**MIRS**; Lactoscope FTIR, Delta Instruments, Drachten, the Netherlands) to measure the composition of retentate and permeate. The weight of skim milk processed under these conditions was sufficient to remove all the water remaining in the system. At the end of flushing the system with skim milk, the retentate and permeate outlets were put in recirculation back to the feed tank, and about 320 kg of pasteurized skim milk heated up to 50°C was added to the feed tank.

Stage 1. The 320 kg of skim milk was recirculated for 30 min to allow the system to equilibrate. The retentate and permeate removal rates were 1,020 and 2,040 g per 60 s. The target flux was 72 kg/m² per hour. Samples of retentate and permeate were taken for analysis by MIRS, the flows of retentate and permeate were measured (the weight of the liquid collected during 60 s) at 5, 15, and 25 min of recirculation and CF was calculated based on the weights of retentate and permeate collected during 60 s. The target CF was 3.00 $\pm$ 0.05. If the CF was below or above the targeted value the retentate or permeate removal rates were adjusted. The Hunter L, a, b color values were measured for

permeates during recirculation as an indication of the clarity of the permeates. After 30 min of recirculation, the retentate and permeate were collected. The typical Rpi and Ppi were about 460.6 and 245.6 kPa at the top end of the vertical membrane, respectively, and the Rpo and Ppo were 230.0 and 259.4 kPa, respectively, at the bottom end of the vertical membrane. Every 15 min, CF was calculated based on retentate and permeate collected in 60 s, permeate flux (kg/m^2 per hour) was determined as the weight of the permeate collected in 60 s, and samples of permeate and retentate were taken for analysis by MIRS to monitor the composition of the retentate and permeate. When the feed tank was nearly empty, the retentate and permeate outlets were transferred to the feed vat for recirculation while stage 1 retentate and RO water were added to the feed vat for the second stage of processing. At the end of stage 1, all of the retentate and permeate were collected, mixed separately, and sampled. The mean CF during stage 1 was 2.96. The average total time of milk processing during stage 1 was about 104 min.

Stage 2. The final retentate (105 kg) from stage 1 was mixed with pasteurized RO water at 50°C at a ratio of 1:2 and transferred to the feed tank. The material was recirculated for about 20 min. At the start of the 20 min of recirculation, the permeate and retentate removal rates were adjusted to 2,320 and 1,160 g in 60 s., which was approximately 140 and 70 L/h on the flow meters, respectively. The target flux for stage 2 was 82 kg/m^2 per hour. The samples of the retentate and permeate were taken for analysis by MIRS at 5 and 10 min of recirculation. The removal rates of retentate and permeate were measured and CF was calculated. If the CF was not in the range of 3.0 ± 0.05 the removal rates of permeate or retentate were adjusted. At the end of 20 min of recirculation, retentate and permeate were collected. Retentate and permeate removal rates were measured every 15 min as a weight of liquid collected in 60 s and CF was calculated together with permeate flux. The samples of retentate and permeate were taken for analysis at 15-min intervals. The typical pressures were 449.4 (Rpi) and 245.4 (Ppi) and 230.0 (Rpo) and 259.2 kPa (Ppo) at the top and bottom end of the vertical membrane, respectively. When the feed tank was nearly empty, the retentate and permeate outlets were transferred to the feed tank for recirculation. The retentate and permeate were mixed separately and each was sampled. The average processing time was 89 min and CF was 2.99.

Stage 3. The final retentate (104 kg) from Stage 2 was mixed with pasteurized RO water at a ratio of 1:2 and transfer to the feed tank. The mixture was recirculated for 20 min. At the beginning of 20 min of recirculation,

the retentate and permeate removal rates were adjusted to 2,600 and 1,300 g in 60s., which was approximately 156 and 78 L/h on the flowmeters, respectively. The target flux for stage 2 was 92 kg/m^2 per hour. At 5 and 10 min of recirculation, the samples of retentate and permeate (samples taken from the outlets) were taken for analysis by MIRS, the removal rates were measured, and CF and permeate flux were calculated. The target CF was 3.0 ± 0.05 . If CF was not in that range, the removal rates were adjusted and new values were recorded. The permeate samples during recirculation were analyzed for Hunter L, a, b color values. At the end of 20 min of recirculation, retentate and permeate were collected. In 15-min intervals, the samples of retentate and permeate were analyzed, removal rates were measured as a weight of the liquid collected in 60 s, and CF and permeate flux were calculated. If needed, the retentate and permeate removal rates were adjusted to achieve CF of 3.0 ± 0.05 . The typical Rpi and Ppi were about 443.1 and 244.0 kPa at the top end of the vertical membrane, respectively, and the Rpo and Ppo were 230.0 and 257.8 kPa, respectively, at the bottom end of the vertical membrane. When the feed tank was nearly empty, the retentate and permeate outlets were transferred to the feed tank. The retentate and permeate were mixed separately and each was sampled. The average processing time was 78 min and CF was 3.03. The cleaning procedure applied after processing (same as described by Zulewska et al., 2009) enabled us to restore initial water flux before processing.

Chemical and Instrumental Analyses

The raw and pasteurized skim milks (samples taken directly before and after pasteurization, respectively) were analyzed for total N (TN), noncasein N (NCN), and NPN content using Kjeldahl methods from AOAC International (2000; method 991.20; 33.2.11; method 998.05; 33.2.64; and method 991.21; 33.2.12, respectively). Skim milks, permeates, and retentates collected during processing were analyzed using MIRS (Lactoscope FTIR, Delta Instruments) for fat, lactose, and true protein content (Kaylegian et al., 2006). This was done to monitor the composition of retentate and permeate during the run to detect if the system was running normally.

Skim milk and final retentates (stages 1, 2, and 3) were analyzed for TS, TN, NCN, NPN, and lactose content using forced-air oven drying (AOAC International, 2000; method 990.20; 33.2.44), Kjeldahl (AOAC International, 2000; method 991.20; 33.2.11), Kjeldahl (AOAC International, 2000; method 998.05; 33.2.64), Kjeldahl (AOAC International, 2000; method 991.21;

Table 1. Mean ($n = 3$) composition (% by weight) of raw and pasteurized skim milks (samples taken directly before and after pasteurization)

Milk treatment	Composition ¹						Serum protein
	CP	NCN	NPN	TP	CN	CN%TP	
Raw	3.25 ^a	0.74 ^a	0.20 ^a	3.06 ^a	2.51 ^a	82.07 ^b	0.55 ^a
Pasteurized	3.25 ^a	0.72 ^b	0.19 ^b	3.07 ^a	2.53 ^a	82.57 ^a	0.53 ^b
R ²	0.91	0.88	0.92	0.84	0.88	0.88	0.82
SE	0.006	0.002	0.002	0.007	0.007	0.08	0.003

^{a,b}Means in the same column not sharing a common superscript are different ($P < 0.05$).

¹CP = total N $\times$ 6.38; NCN = noncasein nitrogen $\times$ 6.38; NPN = nonprotein nitrogen $\times$ 6.38; TP = true protein (CP – NPN); CN = CP – NCN; CN%TP = casein as a percentage of true protein; serum protein = (TP – casein).

33.2.12), and an enzymatic lactose method (AOAC International, 2000; method 2006.06, 33.2.67), respectively. Permeates (stages 1, 2, and 3) were analyzed for TS, TN, NCN, and NPN using the methods indicated above. The TN, NCN, and NPN were multiplied by 6.38 to express results on a protein basis. True protein was calculated by subtracting NPN from TN, CN was calculated by subtracting the NCN from TN, and SP content was calculated by subtracting NPN from NCN. Skim milks and final retentates from stage 3 were analyzed for fat content using ether extraction (AOAC International, 2000; method 989.05; 33.2.26). All samples were analyzed fresh. The SP content in the permeate portion of the skim milk (expressed as a percentage) was calculated by dividing SP in milk by SP in the permeate portion of the milk multiplied by 100, where the permeate portion of milk is considered as 100 kg minus the weight of fat plus CN in 100 kg of milk.

Determination of Percent SP Removal and SP Removal Rate

The efficiency of removal of SP from skim milk was measured in a mass-balance experiment by measuring the weight of skim milk to be processed and the concentration of SP in the skim milk measured by Kjeldahl. The total weight of SP present in the skim milk was calculated. The SP removed from the skim milk was calculated by multiplying the weight of permeate removed by the MF process by the concentration of SP in the permeate as measured by Kjeldahl. From those values, the total weight of SP removed was calculated. The weight of SP removed divided by the weight of SP at the beginning multiplied by 100 is the percent SP reduction achieved by the MF process. The SP removal rate (kg/m² per hour) was calculated by dividing SP removed from the skim milk (SP in the permeate) by membrane surface area (1.7 m²), then SP removed by meters-squared of the membrane was multiplied by 60 min and divided by processing time.

RESULTS

Composition of Skim Milk

The time and temperature of pasteurization (72°C, 16 s) caused a small but detectable ($P < 0.05$) increase in the apparent CN as a percentage of true protein (Table 1) as a result of thermal denaturation of β -lactoglobulin (BLG) and its bonding to κ -CN, as described by Lynch and Barbano (1998). The BLG and κ -CN interaction caused a decrease in the SP content of pasteurized milk ($P > 0.05$; Table 1). Hurt and Barbano (2010) demonstrated that increased heat treatment of skim milk caused a decrease in SP removal and, thus, in the yield of milk SP isolate. In current study, the parameters of pasteurization were chosen to minimize the degree of BLG bonding to κ -CN.

Operational Parameters

Mean operational pressures recorded during 3 stages of MF process using 0.1- μ m GP membranes are shown in Table 2. Differences were detected ($P < 0.05$) in TMP at the TMPin among stages, with stage 1 having the highest TMPin. No difference ($P > 0.05$) was detected in TMPin between stages 2 and 3. The decrease in TMPin, calculated as Rpi minus Ppi, was a function of retentate pressure at the inlet. The Ppi was stable during 3 stages; no difference was detected ($P > 0.05$) among stages. The decrease in Rpi, and thus TMPin, may be a result of the progressive change (i.e., decrease) in viscosity of feed from stage to stage, as retentate produced by stage 1 was diluted with RO water and was used as the feed for stage 2, and so on.

The decrease in lactose, soluble mineral, and SP content of the retentate that was diluted with water and DF, resulted in lower viscosity of the feed material in subsequent stages. Lower viscosity of the recirculating retentate caused a decrease in Rpi, thus the decrease in TMPin was probably caused by changes in the com-

Table 2. Mean ($n = 3$) operational pressures recorded during 3 stages of a microfiltration with diafiltration of skim milk using 0.1- μm ceramic graded permeability (GP) membranes

Stage of processing (kPa)	Operational parameter ¹						
	Rpi	Ppi	TMPin	Rpo	Ppo	TMPout	ΔTMP
1	460.64 ^a	245.57 ^a	230.74 ^a	230.00	259.36 ^a	−26.90 ^a	257.64 ^a
2	449.38 ^b	245.43 ^a	219.62 ^b	230.00	259.22 ^a	−26.76 ^a	246.38 ^a
3	443.13 ^c	243.97 ^a	214.83 ^b	230.00	257.76 ^a	−25.30 ^a	247.10 ^a
R ²	>0.99	0.68	0.95	—	0.68	0.42	0.71
SE	0.41	1.57	1.48	—	1.57	1.57	4.40

^{a–c}Means in the same column not sharing a common superscript are different ($P < 0.05$).

¹Rpi = retentate pressure inlet; Ppi = permeate pressure inlet; TMPin (transmembrane pressure inlet) = Rpi – Ppi; Rpo = retentate pressure outlet; Ppo = permeate pressure outlet; TMPout (transmembrane pressure outlet) = Rpo – Ppo; ΔTMP = TMPin – TMPout.

position of the feed material with increasing stages of processing.

No differences ($P > 0.05$) were detected in pressures at TMPout for both retentate and permeate. Negative values of TMPout (Table 2) indicate a Starling flow was present at the bottom end of the membrane. The concept of Starling flow was described by Heath et al. (1990), and was also reported in our previous work with the GP membranes (Zulewska et al., 2009). The removal rates of permeate and retentate were increased from stage (2,040 and 1,020, 2,320 and 1,160, and 2,600 and 1,300 g/min for stages 1, 2, and 3, respectively), but this still caused a Starling flow. The pressures profiles were stable across time of operation within each stage (Figure 1a and b). The total processing time in this experiment was almost 7 h; however, we were able to maintain stable TMP for each stage (Figure 1a and b; Table 3). A small ($P < 0.05$) decrease was observed in TMPin and TMPin – TMPout with time of processing and this decrease was different depending on stage (i.e., time $\times$ stage interaction); however, the amount of total variation explained by these terms in the ANOVA model was small (Table 3). Therefore, very little increase in fouling occurred during the 7 h of processing.

The retentate recirculation rate and linear velocity was lower ($P < 0.05$) for stage 1 than for stages 2 and 3 (Table 4). No differences ($P > 0.05$) in retentate recirculation rate and linear velocity were detected between stages 2 and 3 (Table 4). Retentate recirculation rate (660 L/min) and linear velocity (6.6 m/s) measured for the UTP system were significantly lower than GP within each stage. The UTP system was designed to operate at average processing flux of about 54 L/m² per hour to maintain a low and uniform TMP from membrane inlet to outlet. It may be possible to operate a UTP MF system at flux >54 L/m² per hour, but no data has been reported on operation of a UTP MF system at flux >54 L/m² per hour. The GP membranes allow operation at higher flux, thus higher recircula-

tion rate and cross-flow velocity are needed than in the UTP system.

CF

Controlling CF based only on the volumetric flow meters (571–123 and 551–123, for permeate and retentate, respectively; Gemü Valves Inc., Atlanta, GA) did not give adequate control of CF ($3\times$ CF) in previous experiments carried out in our group. To stay within

Figure 1. Mean ($n = 3$) values of transmembrane pressure (a) at the retentate inlet end of the membrane and (b) at the retentate outlet end of the membrane for stages 1, 2, and 3 during processing of skim milk using 0.1- μm ceramic graded permeability (GP) membranes.

Table 3. Type III sum of squares for ANOVA to determine the effect of replicate (Rep), stage of processing (Stage) and time of processing (Time) on transmembrane pressure inlet (TMPin), transmembrane pressure outlet (TMPout), and Δ TMP (TMPin – TMPout)

Source	df	TMPin	TMPout	Δ TMP
Rep	2	862.08*	91.59*	398.56*
Stage	2	2,611.58*	32.88*	2,960.65*
Rep $\times$ stage	4	165.93*	173.71*	20.03*
Time	1	98.83*	6.08	153.96*
Time $\times$ rep	2	72.98*	33.64*	16.55*
Time $\times$ stage	2	94.73*	20.61*	26.99*
Model error	44	135.92	96.35	45.02
R ²		0.97	0.80	0.99

* = $P < 0.05$.

the control limits of 3 ± 0.05 for CF, the retentate and permeate removal rates were adjusted based on CF calculated from the weights of retentate and permeate collected in 60 s. No differences ($P > 0.05$) in CF were detected among stages (Table 5). Adjusting CF based on the weights of retentate and permeate during processing gave better control of final CF than using volumetric flow meters. For each stage, control of CF was well within the target range ($\pm 0.05\times$). Controlling CF has a large influence on composition of the retentate and SP removal (Hurt and Barbano, 2010).

Flux

Flux was stable through the time of run within each stage (Figure 2). No difference ($P > 0.05$) in flux for each stage was detected with time as a continuous variable (Table 6). During 3-stage MF process, GP was able to achieve a higher ($P < 0.05$) flux than UTP within each stage (Table 7). The UTP system was operated at average flux of 54 kg/m² per hour, which was recommended by the manufacturer of the membranes. In the present study, the GP system was operated at higher flux than the previous study (71.79 kg/m² per hour) using GP membranes (Zulewska et al., 2009) and

higher cross-flow velocity. Flux was progressively increased with increasing stage, resulting in average flux of 72.49, 84.53, and 92.66 kg/m² per hour for stages 1, 2, and 3, respectively. No cleaning of the membranes occurred between stages, so the fact that we were able to progressively increase flux during milk processing from one stage to the next indicated that no buildup of foulant on the membrane occurred to limit permeability of the membrane during the 7 h of running time at 50°C and demonstrates the capability of the membrane to perform well at higher flux, so long as certain parameters are well controlled during the run. Permeation flux and high cross-flow velocity are the most important factors in limiting the fouling and contributing to good selectivity of the membrane and will be further discussed in this paper.

The degree of fouling was measured based on water permeation fluxes before and after processing skim milk. The process consisted of 3 subsequent stages and we did not clean the system between the stages. Fouled water flux measured directly after processing of skim milk with GP membranes corresponded to 68% of the initial clean water flux (almost 32% decrease) before processing (Table 8). The initial clean water flux of 0.1- μ m GP membranes was about 40% of that of the UTP

Table 4. Mean ($n = 3$) retentate recirculation rate (L/min) and recirculation linear velocity (m/s) recorded during 3 stages of a microfiltration with diafiltration of skim milk using 0.1- μ m ceramic graded permeability (GP) membranes

Stage of processing	Retentate recirculation rate (L/min)	Recirculation linear velocity (m/s)	
		GP	UTP ¹
1	712.59 ^b	7.11 ^b	6.60
2	713.72 ^a	7.12 ^a	6.60
3	714.16 ^a	7.13 ^a	6.60
R ²	0.95	0.95	
SE	0.13	0.001	

^{a,b}Means in the same column not sharing a common superscript are different ($P < 0.05$).

¹Data from our previous work with uniform transmembrane pressure (UTP) system when processing skim milk at 50°C, flux at 53.68 kg/m² per hour, concentration factor 3 $\times$, and recirculation rate at 660.82 L/min. Stage 2 and 3 were assumed to have the same linear velocity as stage 1 because the conditions during processing did not change.

Figure 2. Mean ($n = 3$) flux in different stages of microfiltration during time of processing using 0.1- μm ceramic graded permeability membranes.

membranes (Table 8) when measured under the same conditions in the same pilot-scale system, indicating the higher hydraulic resistance of the GP membrane structure than the UFP membrane structure. The clean and fouled membrane water flux data reported in an experiment using UTP membranes (Hurt et al., 2010) decreased by approximately 41, 25, and 21% in stages 1, 2, and 3, respectively (Table 8), indicating that fouling of UTP membranes decreased as lactose, soluble minerals, and SP were removed. In the case of processing skim milk with UTP membranes, the system was cleaned between each stage. The UTP system was operated with lower cross-flow velocity than the GP system (Table 4), thus lower shear stress. The wall shear stress ($\tau_w = f\rho v^2/2$) was calculated from measured cross-flow velocities (v) using Blasius's law (friction factor, $f = 0.0792 \text{ Re}^{-0.25}$; Samuelsson et al., 1997). In our calculations, we used a density (ρ) of skim milk of 1,018.2 kg/m³, viscosity of 0.00075 Pa/s, and inner diameter of the

Table 5. Mean ($n = 3$) concentration factor during 3 stages of microfiltration with diafiltration of skim milk using 0.1- μm ceramic graded permeability (GP) membranes

Stage of processing	Concentration factor	
	Final weight ¹	Partial weight ²
Stage 1	2.96 ^a	2.96 ^a
Stage 2	2.99 ^a	2.99 ^a
Stage 3	3.03 ^a	3.02 ^a
R ²	0.69	0.79
SE	0.02	0.01

^aMeans in the same column not sharing a common superscript are different ($P < 0.05$).

¹Concentration factor determined based on total weights of retentate and permeate collected in each stage.

²Concentration factor determined based on the weights of permeate and retentate collected in 60 s.

Table 6. Type III sum of squares for ANOVA to determine the effect of replicate (Rep), stage of processing (Stage), and time of processing (Time) on flux

Source	df	Flux
Rep	2	1.31
Stage	2	4,282.62*
Rep $\times$ stage	4	32.60
Time	1	7.17
Time $\times$ rep	2	5.72
Time $\times$ stage	2	10.77
Model error	67	392.44
R ²		0.94

* = $P < 0.05$.

tube of 0.004 m; the wall shear stress was 122.9 and 146 N·m⁻² for UTP and GP membranes, respectively.

Higher flux indicates that less membrane surface area is needed to process the same amount of milk in the same length of time. At the operational conditions applied (Hurt et al., 2010), a UTP system would require 2,051 m² of membrane surface to process 1 million kg of milk in 18 h of processing, which is 34% more surface than GP system (Table 9). If it would be possible to run the GP system with a flux of 92 kg/m² per hour for each of 3 stages the membrane, surface area needed to process 1 million kg of milk in 18 h would be 70% less than UTP (Table 9). Operating at higher flux improves the economic feasibility of the MF process because of lower membrane surface area requirements. Less membrane surface area means not only savings on investment costs, but also less energy requirements and less water and chemicals to clean the MF system. Further work is necessary to examine the potential of UTP membranes to operate at higher flux.

Composition of Retentates

The composition of the retentates from stages 1, 2, and 3 are presented in Table 10. Total solids content decreased from stage 1 to 3, due to a decrease in lactose, soluble mineral, and protein content. Diafiltration removed a large portion of the lactose due to dilution of the retentate with water and passage of lactose through the membrane. Crude protein content decreased significantly ($P < 0.05$) as a result of decrease in NPN and SP content. No differences were detected ($P > 0.05$) in CN content for retentates from stages 1, 2, and 3. Controlling CF resulted in constant CN content in the retentate, as predicted by Hurt and Barbano (2010).

Fat content of the final retentate was higher than the skim milk used in this study as a result of fat retention by the membrane (Table 10). Casein as a percentage of true protein was about 95%, and the remaining ca. 5% were assumed to be SP. However, this could be an overestimate given the limitations of using the NCN

Table 7. Mean ($n = 3$) flux (kg/m^2 per hour) during 3 stages of microfiltration with diafiltration of skim milk using $0.1\text{-}\mu\text{m}$ ceramic graded permeability (GP) and ceramic uniform transmembrane pressure (UTP) membranes

Stage of processing	Flux ¹		R^2	SE	
	GP	UTP ²		GP	UTP
Stage 1	72.49 ^a	53.96 ^b	>0.99	0.37	0.32
Stage 2	84.53 ^a	53.96 ^b	>0.99	0.68	0.59
Stage 3	92.66 ^a	54.62 ^b	>0.99	0.29	0.26

^{a,b}Means in the same row not sharing a common superscript are different ($P < 0.05$).¹Flux was measured based on the weight of permeate collected in 60 s.²Data from Hurt et al. (2010).**Table 8.** Mean water flux during 3 stages of microfiltration with diafiltration of skim milk using $0.1\text{-}\mu\text{m}$ ceramic graded permeability (GP; $n = 3$) and ceramic uniform transmembrane pressure (UTP; $n = 3$) membranes

Stage of processing	Clean water flux after initial cleaning and before processing (kg/m^2 per hour)				Fouled water flux after processing skim milk (kg/m^2 per hour)				Clean water flux after cleaning postprocessing (kg/m^2 per hour)			
	GP	SD	UTP ¹	SD	GP	SD	UTP ¹	SD	GP	SD	UTP ¹	SD
Stage 1	403.9	3.4	1,054.4	10.1	—	—	626.5	151.3	—	—	1,054.4	5.6
Stage 2	—	—	1,057.4	2.9	—	—	796.3	50.6	—	—	1,051.0	6.8
Stage 3	—	—	1,051.5	11.1	276.5	10.2	831.4	39.2	417.7	10.2	1,058.8	10.7

¹Data from Hurt et al. (2010).**Table 9.** Calculated membrane surface area (m^2) for $0.1\text{-}\mu\text{m}$ ceramic graded permeability (GP) and uniform transmembrane pressure (UTP) membranes required to process 1 million kg of skim milk in an 18-h run based on different permeate flux (kg/m^2 per hour)

Item	GP				UTP			
	Actual flux ¹		92 kg/m^2 per hour for all stages		Actual flux ¹			
Stage 1	511		403		686			
Stage 2	438		403		686			
Stage 3	400		403		678			
Total	1,349		1,208		2,051			

¹Calculated based on flux data in Table 6.**Table 10.** Mean ($n = 3$) composition (% by weight) of skim milk (samples taken directly from feed vat before processing) and retentates produced using $0.1\text{-}\mu\text{m}$ ceramic graded permeability (GP) membranes

Item	Composition ¹										SP in permeate portion
	TS	Fat	Lactose	CP	NCN	NPN	TP	CN	CN%TP	SP	
Milk	9.13	0.10	4.76	3.26	0.74	0.20	3.06	2.52	82.44	0.54	0.55
Retentate											
Stage 1	14.23 ^a	na ²	4.41 ^a	8.01 ^a	0.96 ^a	0.18 ^a	7.83 ^a	7.05 ^a	90.06 ^c	0.78 ^a	0.26
Stage 2	10.69 ^b	na	1.69 ^b	7.58 ^b	0.58 ^b	0.09 ^b	7.49 ^b	7.01 ^a	93.52 ^b	0.49 ^b	0.16
Stage 3	9.34 ^c	0.28	0.66 ^c	7.44 ^c	0.40 ^c	0.05 ^c	7.39 ^c	7.05 ^a	95.35 ^a	0.34 ^c	0.12
R^2	>0.99	—	>0.99	0.94	>0.99	0.99	0.91	0.74	0.99	0.99	—
SE	0.04	—	0.005	0.03	0.005	0.003	0.03	0.03	0.08	0.006	—

^{a-c}Means in the same column not sharing a common superscript are different ($P < 0.05$).¹TS = total solids; CP = total N $\times$ 6.38; NCN = noncasein nitrogen $\times$ 6.38; NPN = nonprotein nitrogen $\times$ 6.38; TP = true protein (CP – NPN); CN = CP – NCN; CN%TP = casein as percentage of true protein; SP = serum proteins (TP – CN).²na = not applicable.

Table 11. Mean ($n = 3$) composition (% by weight) of permeates produced using 0.1- μm ceramic graded permeability (GP) membranes

Stage of processing	Composition ¹							
	TS	CP	NCN	NPN	TP	CN	CN%TP	SP
Stage 1	6.48 ^a	0.73 ^a	0.70 ^a	0.19 ^a	0.54 ^a	0.029 ^a	5.25 ^a	0.51 ^a
Stage 2	2.60 ^b	0.35 ^b	0.32 ^b	0.08 ^b	0.27 ^b	0.026 ^a	9.58 ^a	0.25 ^b
Stage 3	1.08 ^c	0.19 ^c	0.17 ^c	0.04 ^c	0.15 ^c	0.025 ^a	16.36 ^a	0.13 ^c
R ²	>0.99	>0.99	>0.99	0.99	>0.99	0.27	0.80	>0.99
SE	0.005	0.002	0.002	0.002	0.002	0.003	1.13	0.003

^{a-c}Means in the same column not sharing a common superscript are different ($P < 0.05$).

¹CP = total N $\times$ 6.38; NCN = noncasein nitrogen $\times$ 6.38; NPN = nonprotein nitrogen $\times$ 6.38; TP = true protein (CP – NPN); CN = CP – NCN; CN%TP = casein as percentage of true protein; SP = serum proteins (TP – CN).

method on MF retentates (Nelson and Barbano, 2005). Low content of SP and lactose in the final product may produce MF retentate with higher thermal stability, as a result of removing SP, which are much more temperature sensitive than CN (Anema, 2009). Lactose also undergoes thermal degradation, including Maillard reactions with proteins that can lead to off flavors and browning (Walstra et al., 1999). Moreover, low lactose level presents additional benefits in application of micellar CN concentrate in products for diabetics or people with low lactose tolerance.

Composition of Permeates

The TS, CP, NCN, and NPN content in permeate (Table 11) decreased with increasing stage of processing ($P < 0.05$) as a result of SP removal in the previous stage and retentate dilution with water between stages. No difference in CN content of permeates from stages 1, 2, and 3 was detected ($P > 0.05$); the average CN content in permeate for stages 1, 2, and 3 was 0.027%. The SP content in first stage permeate was lower than SP in permeate portion of the pasteurized skim milk, which was calculated at 0.55% (Table 11). This was primarily due to some rejection of SP by the membrane. As calculated using the same calculation method as Hurt and Barbano (2010), stage 2 and 3 permeate should contain 0.26 and 0.16% of SP, respectively (Table 10). In our experiment, the permeate contained 0.25 and 0.13% of SP in second and third stages, respectively, and no difference was detected ($P > 0.05$) between calculated and actual SP content in permeate.

pH

The pH (Table 12) of the starting material and retentate increased with increasing stage ($P < 0.05$). The starting material for stage 1 was skim milk at 50°C, and in subsequent stages the starting materials were the retentates from previous stage diluted with 2 parts pasteurized RO water to 1 part retentate by

weight. Dilution of the retentate with RO water resulted in an increase in pH of the starting material; as a consequence, the retentate produced had higher pH than retentate from previous stage. A higher pH of the micellar CN concentrate may also provide higher heat stability when it is used as an ingredient in retorted beverages (Sauer and Moraru, 2012).

SP Removal

The efficiency of MF process can be measured by removal of SP by the membrane. As reported in our other studies (Zulewska et al., 2009), the type of the membrane can have a great effect on the percentage of SP removal, and thus on the composition and properties of the final products. Theoretical SP removal and data for GP and UTP membranes are shown in Table 13. The UTP membranes removed a higher percentage of the SP available ($P < 0.05$) in stage 1 of the processing; however, a higher percentage SP removal ($P < 0.05$) was observed for GP in stages 2 and 3 than for UTP. Cumulative SP removal for stages 1 and 2 was higher ($P < 0.05$) for UTP system (Table 13), but overall no differences in cumulative SP removal for the 3 stages were detected ($P > 0.05$; Table 13). Both types of membranes can produce a similar final product in terms of SP removed; however, energy requirements for UTP membranes are much higher than GP, which

Table 12. Mean ($n = 3$) pH values (50°C) of the stage starting material and final retentate from each stage of the ceramic graded permeability (GP) system

Stage of processing	Starting material	Retentate
Stage 1	6.51 ^c	6.53 ^c
Stage 2	6.75 ^b	6.73 ^b
Stage 3	6.95 ^a	6.86 ^a
R ²	0.99	0.96
SE	0.02	0.03

^{a-c}Means in the same column not sharing a common superscript are different ($P < 0.05$).

Table 13. Serum protein (SP) removal (%) by stage for 0.1- μm ceramic graded permeability (GP) and 0.1- μm ceramic uniform transmembrane pressure (UTP) membranes

Stage of processing	Removal (%)			R^2	SE	
	Theoretical ¹	GP	UTP ²		GP	UTP
SP						
Stage 1	68.0	55.97 ^b	63.66 ^a	0.98	0.37	0.32
Stage 2	22.0	26.67 ^a	21.92 ^b	0.93	0.43	0.37
Stage 3	7.0	13.81 ^a	9.65 ^b	0.87	0.54	0.46
Cumulative SP						
Stage 1	68.0	55.97 ^b	63.66 ^a	0.98	0.37	0.32
Stage 2	90.0	82.64 ^b	85.59 ^a	0.64	0.75	0.65
Stage 3	97.0	96.45 ^a	95.23 ^a	0.10	1.22	1.05

^{a,b}Means in the same row not sharing a common superscript are different ($P < 0.05$).

¹Assuming no rejection of SP and complete rejection of casein.

²Data from Hurt et al. (2010). Assuming 0.03% of casein in TP of permeate.

increases the overall cost of the process and price of final product. In the UTP system, apart from the feed and retentate pump, a permeate pump circulates the permeate parallel to the membrane surface in the same direction as retentate flow to create a corresponding decrease in pressure on the permeate side of the system. As a result, the pressure difference between both sides of the filter is substantially maintained constant throughout the entire length of the membrane (Sandblom, 1978). The drawback of such an approach is that to achieve a high pressure in the recirculated permeate a large pumping capacity is needed.

SP Passage

Taking into consideration the economics of the MF process, it is very important to calculate the efficiency of SP removal expressed in kilograms of SP removed per unit of membrane surface area per hour within each stage. The data from the current experiment, together with the results for UTP membranes reported by Hurt et al. (2010), are presented in Table 14. The SP removal rate was higher ($P < 0.05$) for GP membranes for each stage as compared with UTP membranes. As a consequence, cumulative SP passage was higher for GP than UTP, at 0.69 versus 0.46 kg/m² per hour, respectively.

Higher SP removal rate for GP membranes was due to higher flux during processing for all 3 stages. We may have been able to increase SP removal rate to 0.82 kg of SP/m² per hour if we ran the system with a flux of 92 kg/m² per hour for all 3 stages. Future work should identify factors limiting membrane performance as the basis for improvement in design of membranes.

DISCUSSION

Several parameters have to be controlled during MF processing to minimize fouling and ensure proper selectivity of the membrane. It must be noted that the pressure on the retentate side of the membrane from the inlet to the outlet of a membrane (i.e., approximately 1 m in length) decreases from the inlet to outlet end. The structure of GP and Isoflux ceramic membranes provides higher hydraulic resistance at the inlet to a lower hydraulic resistance at outlet end of the membrane. The difference in TMP at the inlet versus outlet end of a membrane stick is much larger for the GP and Isoflux membranes than for UTP membranes operated with a permeate recirculation pump. This means that, in reality, a range of critical and limiting fluxes may exist from the inlet to outlet of a ceramic membrane stick. The critical flux is defined as the flux

Table 14. Serum protein (SP) removal rate (kg/m² per hour) for 0.1- μm ceramic graded permeability (GP) and 0.1- μm ceramic uniform transmembrane pressure (UTP) membranes

Stage of processing	SP removal rate		R^2	SE	
	GP	UTP ¹		GP	UTP
Stage 1	0.37 ^a	0.29 ^b	>0.99	0.002	0.001
Stage 2	0.20 ^a	0.11 ^b	0.98	0.005	0.004
Stage 3	0.12 ^a	0.06 ^b	0.95	0.005	0.005
Cumulative	0.69 ^a	0.46 ^b	0.99	0.009	0.008

^{a,b}Means in the same row not sharing a common superscript are different ($P < 0.05$).

¹Data from Hurt et al. (2010).

above which irreversible fouling occurs at each point along the length of the membrane and the limiting flux is the maximum flux that can be achieved at steady state and corresponds to the point where the critical flux has been reached at all points along the length of the membrane (Bacchin, 2004). Overall, the average critical and limiting flux over the full length of the membrane are what are observed and reported. Permeation flux and tangential flow velocity, through the wall shear stress, are the major parameters responsible for a proper membrane-separation process (Le Berre and Daufin, 1996). Transmembrane pressure is a driving force for flux (drives fluid through the membrane carrying along solutes that are soluble in the permeate and that are not rejected by the membrane because of their size). According to Darcy's law, increasing TMP results in higher flux; however, it is only true for pure solvent solutions (Wu et al., 1999). In reality, raising TMP increases the fouling rate, especially by compacting the fouling layer (Gesán-Guizoiu et al., 1999). Therefore, it is crucial to operate the MF process at less flux than the limiting flux. The limiting flux represents the maximum stationary permeation flux that can be reached when increasing TMP (Aimar and Field, 1992; Bacchin 2004) and depends on cross-flow velocity, temperature, feed material properties, and geometry of the membrane module (Samuelsson et al., 1997). Samuelsson et al. (1997) investigated 4 models for rejected solute back-transport mechanisms in cross-flow microfiltration to predict the limiting permeate flux for skim milk. Among the models discussed (i.e., Brownian diffusion model, shear-induced diffusion model, inertial lift model, and surface transport model) by Samuelsson et al. (1997), only shear-induced diffusion model predicted values of the limiting flux close to their experimental data. Samuelsson et al. (1997) proposed the empirical relation $\text{flux} = \text{Re} \times 6.94 \times 10^{-10} \text{ m/s}$ (or, $\text{flux} = \text{Re} \times 0.0025 \text{ L/m}^2 \text{ per hour}$) as the best predictor of the limiting flux. We have calculated the limiting flux value for GP membranes operating at a cross-flow velocity of 7.12 m/s to be 96 L/m² per hour [assuming that density (ρ) of skim milk is 1,018.2 kg/m³, viscosity 0.00075 P·s, and inner diameter of the tube 0.004 m]. This means that during our experiment we operated our MF system below the limiting flux value. The limiting flux for UTP system was 89 L/m² per hour.

Flux has an effect on separation performance (through the critical flux), but does not alter the deposit characteristics, and thus permeability and selectivity (Le Berre and Daufin, 1996). In the current experiment, the flux was stable within each stage, indicating no marked fouling occurred (Figure 2). Additionally, we were able to increase flux from stage to stage and maintain good permeability, which may have been due to higher par-

ticle removal rates compared with particle deposition rates.

Unlike the limiting flux, the critical flux is a concept that describes the flux below which little or no deposition exists on the membrane surface and above which a sharp increase in fouling occurs (Howell, 1995; Bacchin et al., 1995; Gesán-Guizoiu et al., 1999; Bacchin 2004). Bacchin (2004) suggested that the critical flux is equal to two-thirds of the limiting flux. In our study (MF system operated with GP membranes), the critical flux value was calculated to be 64 L/m² per hour. The water flux before and after processing can be used an indicator of membrane fouling, whereas, in current experiment, the fouled water flux after processing was 68% of clean water flux before processing (Table 8). This was a result of fouling layer formation on the membrane surface once membranes were exposed to skim milk flow. The flux within each stage was stable, which means little additional fouling occurred. The increase in flux with increasing stage did not cause any flux fluctuations within each stage. Considering that the critical flux is reached when irreversible fouling occurs locally on the membrane, whereas the limiting flux is reached when the whole membrane surface operates above the critical flux (Bacchin 2004), we have operated under conditions of partial membrane fouling. The value of critical flux obtained during cross-flow filtration is a function of particle size and concentration of the feed solution, surface (membrane-feed) interactions, and hydrodynamic conditions (Wu et al., 1999; Bacchin, 2004). In general, as protein concentration in a solution increases, viscosity of the solution (Walstra et al., 1999) and the probability of protein-protein interactions increase. Concerning hydrodynamics, the critical flux is a function of the cross-flow velocity (Bacchin, 2004). Therefore, the difference in the critical flux between UTP and GP membranes results only from operating under different cross-flow velocities (6.6 vs 7.12 for UTP and GP membranes, respectively). The critical flux value for UTP membranes (two-thirds of the limiting flux) was calculated at 59 L/m² per hour; thus, Hurt et al. (2010), operating with the same pumping system equipped with UTP membranes at the flux of 54 L/m² per hour, did not exceed the critical flux value. This raises the question of whether the fouling was more severe when processing skim milk with GP membranes or with UTP membranes. In Table 8 the water fluxes are presented for both systems before and after processing skim milk. A 41% (59% of the initial clean water flux before processing) decrease was observed in water flux after stage 1 of processing skim milk with UTP membranes, as compared with 32% (68% of the initial flux) with GP membranes (after 3 stages). For UTP stages 2 and 3, a decrease of 25 and 21% in water flux, respec-

tively, was observed (Table 8). The standard cleaning procedure was applied between subsequent stages for system equipped with UTP membranes, whereas no cleaning was done between each stage when processing skim milk with GP membranes. The total processing time for GP membranes was 6.5 h (including recirculating milk between stages), whereas the processing time for UTP system was approximately 8, 7, and 6 h for stages 1, 2, and 3, respectively.

Wall shear stress has both a quantitative and qualitative effect. Increased shear stress results in a decrease of deposit thickness and porosity by preferential movement of larger CN micelles away from the membrane surface and, as a consequence, higher flux. Under certain conditions (flux dependent) a critical value of wall shear stress (τ_w) exists, which causes a decrease in SP passage (Le Berre and Daufin, 1996). Several studies (Le Berre and Daufin, 1996; Gesan-Guizieu et al., 1999, 2000; Grandison et al., 2000) suggested a critical ratio of flux to wall shear stress (convection toward the membrane or erosion) in cross-flow MF, below which no marked fouling by colloidal particles (i.e., CN micelles) exists. Operating an MF system above the critical value of wall shear stress (τ_w) leads to altered performance with a sharp increase in fouling, reduced operating time, a large decrease in permeability, and soluble transmission rates (Gesau-Guizieu et al., 2000). The critical ratio of flux to wall shear stress enables the operator to determine optimal stable filtration operating conditions to ensure high productivity, low processing costs, and minimum cleaning requirements (Gesau-Guizieu et al., 2000). Le Berre and Daufin (1996) assessed the critical value of this ratio close to 1.0 L/h per meter-squared per pascal. When the ratio of flux to wall shear stress is higher than critical value, the convection exceeds the removal of the material, which results in thicker fast-growing deposits with a high porosity. In the opposite situation, a deposit with smaller thickness and lower porosity is formed (Le Berre and Daufin, 1996). In the current experiment, with a wall shear stress of $146 \text{ N}\cdot\text{m}^{-2}$ for GP membranes, the ratio of flux to wall shear stress (τ_w) was 0.50, 0.58, and 0.63, for stages 1, 2, and 3, respectively. Samuelsson et al. (1997) suggested that at wall shear stresses of about $100 \text{ N}\cdot\text{m}^{-2}$, shear-induced diffusion is the dominant mechanism for back transport.

CONCLUSIONS

Microfiltration of skim milk using ceramic UTP and GP membranes resulted in similar final retentates in terms of SP removed. The SP removal rate (expressed by kilograms of SP removed per meter-squared of membrane area) was higher ($P < 0.05$) for GP mem-

branes for each stage versus UTP membranes. Higher passage of SP and SP removal rate for GP membranes, compared with UTP membranes, was achieved by increasing cross-flow velocity during processing of skim milk. Increasing flux in subsequent stages did not affect membrane permeability and fouling. We operated under conditions that produced partial membrane fouling, due to using a flux that was less than limiting flux, but higher than critical flux. Because the critical flux is a function of the cross-flow velocity, the difference in critical flux between UTP and GP membranes of the structure used in the present study resulted only from operating under different cross-flow velocities (6.6 vs 7.12 for UTP and GP membranes, respectively). Conditions that allow MF operation at higher flux will reduce the membrane surface area required to process the same amount of milk in the same length of time. Less membrane surface area reduces investment cost and the amount of energy, water, and chemicals to clean the MF system.

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