Optimization of spiral-wound microfiltration process for production of micellar casein concentrate

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Abstract: Micellar Casein Concentrate (MCC) is manufactured from microfiltration (MF) of skim milk utilizing ceramic or polymeric membrane filtration. While ceramic filtration has higher efficiency, use of polymeric is cost effective and the process is familiar to several US dairy processors. The aim of the present study was to develop an optimized membrane filtration process to produce MCC using spiral wound polymeric membrane filtration (SW MF) system by systematic selection of transmembrane pressure (TMP) and level of diafiltration (DF). Using skim milk as feed material, preliminary lab-scale MF experiments were conducted using 0.5 µm polyvinylidene fluoride (PVDF) membrane. Three TMP (34.5, 62.1, and 103.4 kPa) and three levels of DF (70, 100, and 150%) along with a process without DF as control were used in the study. Effect of TMP and effectiveness of DF on flux rates, SP removal, casein to total protein (CN/TKN) ratio, casein to true protein (CN/TP) ratio, rejection of casein (rej CN) and SP (rej SP) were evaluated. At all TMP values used in the study, the overall flux (O Flux) increased with the level of DF. Highest O Flux of 30.77 liter per meter square per hour (LMH) was obtained with 34.5 kPa pressure and 150% DF. The impact of DF was more pronounced at lower pressures than at the higher pressures used in the study. With controlled DF, instantaneous flux was maintained within 80% of initial flux for the entire process run. For all the experiments, casein has a rejection of 0.97 to 1.0, while serum protein has the lowest rejection of 0.10 at 34.5 kPa pressure and 150% DF level. Use of 34.5 kPa and DF level of 150% contributed to 81.45% SP removal, and casein to true protein ratio of 0.96. SP removal data from the lab-scale experiments were fitted into a mathematical model using DF and square of TMP as factors. The model predicts SP removal within 90-95% of actual SP removal got from the pilot plant experiments.

Keywords: polymeric spiral-wound membrane; microfiltration; transmembrane pressure; diafiltration; micellar casein concentrate

1. Introduction

Separation of different components from a mixture of species is a common requirement in several processing industries. Of the various separation processes, the membrane separation process gained popularity because of several advantages over the other processes. Membrane separation technology was introduced to the dairy processing in the early 1970s as an alternative to some thermal and non-thermal processes [1]. Commonly used membrane processes in the dairy industry are microfiltration (MF), ultrafiltration (UF), nanofiltration (NF) and reverse osmosis (RO). These processes are based on selective permeability of a porous membrane and differ in membrane material, molecular weight cutoff, pore size, and operating pressures. Among these four commonly used processes, MF uses porous membranes with a porosity of 0.1 to 2 µm [2]. MF
is extensively used in defatting of whey stream in production of whey protein isolates, removal of somatic cells, bacteria, spores, and fat globules. There has been increased interest in using the MF in production of micellar casein concentrate (MCC) [3–5]. MCC is produced from MF of skim milk by permeating most of the serum protein (SP) and non-protein nitrogen components, thereby, increasing the ratio of casein to total protein (CN/TKN) and casein to true protein (CN/TP). The retentate obtained from this process is a concentrated colloidal suspension [6] containing casein in micellar form, lactose, minerals, and some serum proteins. MCC has been utilized in some applications, such as cheese making [7], process cheese [8,9], and Greek-style yogurt [10]. Also, MCC has other promising applications, such as nutritional meal replacements, whipped toppings, and coffee whiteners. The permeate obtained from this process is another ideal starting material for manufacturing of native serum protein concentrates, native α-Lactalbumin, and β-Lactoglobulin enriched protein ingredients.

Most of the research on MF of skim milk for MCC production used ceramic MF membranes [5,11–13]. Ceramic membrane systems are capital intensive and membrane replacements are expensive [14]. When compared to these systems, membrane separation systems using polymeric membranes requires less footprint, inexpensive and are familiar with most of the US dairy processors. In recent years, the interest in assessing the suitability and efficiency of polymeric membranes for MCC production has been increased [4,14–18]. It has been shown that using ceramic membranes, over 95% of serum protein could be removed in a 3-stage process in which diafiltration (DF) to a level of 200% (on the feed volume basis) was used. DF is a process in which water is added to the retentate during MF and further concentration is carried out. This step is intended to improve the serum protein (SP) removal and to control the membrane polarization phenomenon [19]. A few studies conducted on the use of polymeric membranes for production of MCC showed that approximately 40% of SP was removed from skim milk at the first stage of MF [4,14] and the cumulative SP removal increased to around 60% in the second stage when 200% of DF water was added to the retentate of the first stage [4]. In this process, skim milk was concentrated to three times (3X) volume reduction (VR) and water equal to the volume of the original milk was added to the retentate. Further filtration was done to a VR of 3X, and this process was repeated twice, totaling to 200% DF. While this shows the positive impact of DF on process efficiency, for maximizing the SP removal, it is essential to retain the membrane permeation characteristics over extended process runs. During MF of skim milk, concentrations of materials retained by the membrane continue to increase with process time. Higher concentration of these materials in the bulk of the fluid leads to more pronounced concentration polarization of membrane or fouling, thereby, negatively impacting the serum protein permeation through the membrane [19]. This effect could be seen when only 40% of SP could be removed during MF without adding DF water, and this percentage could be increased to 70% with DF water. When DF water is added to the retentate, the bulk concentration of material goes down, thereby improving the membrane permeation characteristics. In this process, a controlled DF addition should help minimize the concentration polarization effect and maximize SP removal.

Besides DF, transmembrane pressure (TMP) is also an important operating variable in the MF process. TMP is the driving force in the MF process and of all the membrane processes is the most pressure-sensitive process. Using higher pressure leads to higher initial flux rates but there will be increased concentration polarization or fouling leading to a gradual decrease in flux rates and membrane permeation characteristics [19]. The formation of polarization layer and subsequent compaction of this layer and the membrane at higher pressures lead to a condition of pressure independent operation of MF. To sustain a reasonable flux rate over a longer process run and to maintain membrane permeation characteristics, selection of suitable TMP is very important.
Thus, the objectives of the present study are to evaluate the spiral-wound (SW) MF process for maximizing the SP removal and to select optimum levels of operating pressure and DF.

2. Materials and Methods

2.1. Feed

Fresh skim milk was collected from the South Dakota State University dairy plant. The skim milk was pasteurized at 63 °C for 30 min. For different replications, skim milk collected from different days was utilized.

2.2. Membranes

Polyvinylidene fluoride (PVDF) membranes in a flat sheet configuration (Parker Process Advanced Filtration Division, Oxnard, CA) were used in the lab-scale MF experiments. The membrane sheets were cut to the required size of 14.0 x 15.2 cm for use in OPTISEP membrane unit. All the flat sheet membranes were kept wet with 1% sodium metabisulfite solution and stored at 5 °C, before using. For pilot plant experiments, two 0.5 µm PVDF membranes in SW configuration connected in parallel were used (element model FH 3030-OS03S). Each element is of 97 mm diameter and 762 mm length. The elements have 1.1 mm spacers with a membrane area of 4.3 m² per element.

2.3. Proximate Analysis

Samples of feed, permeate, and retentate were analyzed for total solids (TS) using direct forced air oven drying [20] (AOAC, 2000; method 990.20). Total protein nitrogen (TKN), noncasein nitrogen (NCN) and non-protein nitrogen (NPN) were determined by block digester method using micro Kjeldahl apparatus. The true protein (TP) was calculated as the difference between TKN and NPN. Casein (CN) was calculated as the difference between TKN and NCN. The SP content was calculated as the difference between NCN and NPN. A multiplication factor of 6.38 was used to convert nitrogen to protein.

2.4. SP removal

The SP removal was calculated using micro Kjeldahl data and quantities of skim milk used and permeate collected from the process. The SP removal (%) was calculated by dividing the SP content of permeate (g) by SP content (g) of original skim milk and multiplying by 100.

2.5. Overall Flux

Overall flux (O Flux) was calculated as permeate flow rate per unit filtration area per unit time and is expressed as liters per meter square per hour (LMH).

\[
\text{Overall flux, LMH} = \frac{\text{Permeate flow}[\text{ml}] \times 60}{\text{Collection time}[\text{min}] \times \text{Area of membrane}[\text{m}^2] \times 1000}
\]

In the present study, O Flux was averaged over the entire process time. The feed volume and VR used in the study resulted in a process time of about 2 to 6 h.

2.6. Rejection coefficients for SP and CN

Classical rejection coefficients are generally calculated as the ratio of concentration (C) of any component in permeate (p) and retentate (r). With the processes using DF water, the concentration of any component in the permeate goes down due to the dilution effect. This gives a negative rejection coefficient for that component. To overcome this situation, in the present study rejection coefficients were calculated differently. The pore size of the membrane used in the experiments was 0.5 µ. Theoretically, all the components present in the milk should pass through the membrane resulting in 0 rejec-
tions [19]. This corresponds to an equal concentration of the component both in permeate and retentate. In the present study, rejection of a component is expressed as:

\[ \text{Rej} = 1 - \frac{C_p}{C_f} \]

Where, \( C_p \) is the concentration of any component in the permeate while \( C_f \) is the concentration of the component in the feed after adjusting for the level of DF water. For example, if skim milk has 0.54% serum protein and if an experimental run uses 100% diafiltration, the adjusted concentration of serum protein in feed (\( C_f \)) will be \( 0.54/2 = 0.27\% \).

2.7. Operating Variables

Transmembrane pressure (TMP). TMP is the driving force in MF of skim milk. As discussed in the preceding sections, the magnitude of TMP affects the nature of the pseudo filtration layer (concentration polarization layer) formed on the membrane surface. A higher TMP leads to more compaction of this layer, which impacts the membrane permeation characteristics. To study the effect of TMP, experiments were conducted at three levels of TMP (34.5, 62.1 and 103.4 kPa). The TMP was read using an Ashcroft industrial Duralife pressure gauge with a measuring range of 0-413.7 kPa.

TMP is the difference between the average of inlet and outlet pressures minus the permeate pressure.

\[ \text{TMP} = \frac{P_{\text{in}} + P_{\text{out}}}{2} - P_p \]

Diafiltration (DF). DF is often used in the MF process to maximize the protein recovery in defatting of whey streams and in MF of skim milk for maximizing the removal of SP so that CN to TP ratio in MCC could be increased. In the present study, three levels of DF viz. 70, 100 and 150% (based on the feed volume) were used. A control run without addition of any DF was also used to compare the effectiveness of DF in improving the efficiency of MF process.

**Figure 1.** Schematic diagram of the lab-scale membrane separation process.

1- Feed tank; 2- Pump; 3- Flat sheet unit; 4- Membrane; 5- Permeate collection tank; 6- Weighing balance.

2.8. Experimental Procedure

Lab-scale studies. MF experiments were conducted using a lab-scale plate-and-frame unit (OPTISEP 400-unit part # 20-000-1000) procured from NCSRT, NC. The unit used flat sheet membranes of 14 x 15.2 cm size with a filtration area of 0.02 m². The gasket provided a channel height of 0.5 mm for the feed channel. The TMP was
measured using the Ashcroft industrial Duralife pressure gauge (Ascroft, Stratford, CT06614, measuring range of 0-413.7 kPa) fitted to the end plate. The pressure was varied by controlling the back-pressure valve provided on the retentate line at the outlet of the unit. A variable speed peristaltic pump (Masterflex peristaltic pump, Cat. # EW-77521-40, Cole Parmer, IL, USA) coupled to a standard L/S pump head (Cat # C-07024-21) was used to supply the feed to the membrane unit. The feed flow rate to the membrane unit was maintained at 1.7 L/min. Flat sheet membranes were cut into pieces of 14 x 15.2 cm and were assembled in the filtration unit as per the instruction manual supplied with the filtration unit. Before each run, the membrane was flushed with 6 L of deionized water to flush out the storage solvent. Each batch was started with a feed volume of 800 ml and separation was done on continuous concentration mode as shown in Figure 1, to a final retentate volume of 200 ml, giving a VR of 4. All the experiments were conducted at a temperature of 24 oC and a pH of 6.6.

2.9. Statistical Analysis

All the data were analyzed by One-Way ANOVA to test for significant differences among the treatments with Type I error rate (α) of 0.05 using MINITAB® 19 (Minitab, LLC, Chicago, IL).

3. Results and discussion

3.1. Flux

The data on O flux obtained at all levels of DF and for all the pressures used in the study are presented in Figure 2. Both TMP and DF used in the study had a significant effect on O Flux. Highest O Flux of 23.61 LMH was obtained at the lowest TMP used in the study. As the TMP increased, O Flux decreased. However, O Flux obtained at 34.5 and 62.1 kPa TMP (23.61 and 21.1 LMH, respectively) were statistically not different (P > 0.05). It has been reported that use of 50 kPa TMP, while manufacturing casein concentrate utilizing polymeric membranes did not affect the flux significantly [16], which is similar to the results obtained in this study. As shown in Figure 2, addition of DF water had a positive impact on the O Flux. O flux increased by 50% from 0 to 70% DF at 34.5 kPa TMP, while the increase at the same TMP was about 100% from 0 to 150% DF. For the control run (0% DF), there was no significant difference observed in O Flux for all the pressures used in the study and was about 14.75±0.30 LMH. This shows that for optimum operation of MF, it is essential to select appropriate pressure and DF level. From the data presented, it is also clear that at higher operating pressures, DF becomes less effective in influencing the process flux rates. The lowest flux rates obtained in control runs at all the three pressures may be due to the fact that as more and more concentration of rejected solid build up in the retentate, polarization of membrane becomes more pronounced and dictate the permeation characteristics of the membrane, making the process to operate in pressure independent region. DF showed a considerable influence on the process flux rates, especially at lower pressures used in the study.
Overall flux means (n=3) obtained from the lab-scale studies. All experiments were conducted at 24°C and 800 ml feed was concentrated to a final volume of 200 ml resulting in 4 times volume reduction (VR). DF is diafiltration water added, measured as a percentage of original feed volume.

Means not sharing the same alphabet are significantly different (P < 0.05).

Figure 2. Overall flux means (n=3) obtained from the lab-scale studies. All experiments were conducted at 24°C and 800 ml feed was concentrated to a final volume of 200 ml resulting in 4 times volume reduction (VR). DF is diafiltration water added, measured as a percentage of original feed volume. 

Instantaneous flux rates obtained during the lab-scale run for 34.5 kPa pressure at all DF levels are presented in Figure 3. From the data presented in Figure 3, in case of control run the flux values continued to drop over the entire process run duration of about 2 h. Zulewska et al. reported that the flux of SW system decreased with increasing time of processing [14]. For the other runs, DF water was added at VR of 1.23, 1.45, 1.68, 1.88, 1.88 and 2.67. After the final addition at a VR of 2.67, the product was concentrated till a VR of 4. The valleys and peaks in the flux graphs are due to addition of DF water at different intervals. For control run, the initial flux is about 19.5 LMH and by the end of 3 VR (a concentration factor of 3), the flux was only 60% of the initial flux. For all the DF runs, the flux at 3 VR is about 78-80% of the initial flux. The drop in flux from initial to about 90-95% of the process run time is about 5-10%. The addition of DF in small quantities over several intervals of process run helped sustain the process flux rate for a longer time during the process. Also, the level of DF had a strong effect on the flux rates, resulting in higher flux rates with higher DF level.
Figure 3. Instantaneous flux obtained from the lab-scale studies at 34.5 kPa pressure. Instantaneous fluxes are arrived at by measuring the permeate at various time intervals over the entire process run and averaging for that time interval. All the experiments were conducted at 24°C and 800 ml feed was concentrated to a final volume of 200 ml resulting in 4 volume reduction (VR). DF is diafiltration water percentage based on original feed volume.

Data presented in Figure 4 shows the impact of operating pressure on flux rates. From the data presented, operating pressure showed a strong impact on the flux. The flux at all the time intervals is about 50% higher for process run that used 34.5 kPa pressure when compared to the run that used 103.4 kPa pressure. As highlighted in the earlier sections, MF is pressure sensitive process. Higher pressures promote a more pronounced polarization of membrane. Further, there will be a compaction of this layer and the membrane leading to reduced flux. In case of 103.4 kPa pressure, these phenomena may be overshadowing the advantage of DF, thereby maintaining the difference in flux among the three pressures used in the study.
Figure 4. Instantaneous flux obtained from the lab-scale studies at three pressures used. Diafiltration was 150% of feed volume and was added at 6 intervals. Instantaneous fluxes are arrived at by measuring the permeate at various time intervals over the entire process run and averaging for that time interval. All the experiments were conducted at 24°C and 800 ml feed was concentrated to a final volume of 200 ml resulting in 4 volume reduction (VR). DF is diafiltration the percentage is based on original feed volume.

3.2. SP removal

Serum protein removal data from the experiments are presented in Figure 5. Without DF, SP removal ranged from 35 to 50%, the highest being for 34.5 kPa and the lowest being for 106.4 kPa pressure. Statistical analysis of data showed that the SP removal obtained at 34.5 and 62.1 kPa TMP at 0, 70 and 100% DF are not statistically different (P > 0.05). At all the levels of DF, SP removal decreased with increasing the operating pressure. As discussed in the preceding sections, concentration polarization and compaction of polarized layer, as well as the membrane, may determine the membrane permeation characteristics [15,21] and masking the beneficial effect of DF at higher operating pressures. Use of 70% DF at 34.5 kPa gave same SP removal rate as that of 100% DF at 62.1 kPa pressure. Also, 100% DF at 34.5 kPa pressure gave similar SP removal rate as compared to 150% DF at 62.1kPa pressure. The highest SP removal of 81.45% was obtained with the use of 34.5 kPa pressure and 150% DF. The results highlight the importance of selection of appropriate operating pressure and level of DF for maximizing SP removal.

Figure 5. Data on serum protein (SP) removal obtained from the lab-scale studies at three pressures and for all levels of diafiltration used. Diafiltration (DF) water was on feed volume basis and was added at 6 intervals during the process. All the experiments were conducted at 24°C and 800 ml feed was concentrated to a final volume of 200 ml resulting in 4 volume reduction (VR).

The SP removal data obtained from the lab-scale runs were fitted into a mathematical model and expressed as a function of DF level and square of TMP:

\[
SP \% = 58.705 - 0.0015 \times TMP^2 + 0.18 \times DF
\]

Where TMP is in kPa and DF is % of water added based on feed volume. Using the model, it is possible to predict SP removal at any selected operating conditions.
3.3. Rejection of SP and CN

The data on CN/TKN, CN/TP ratios, rejection of CN (rej CN) and SP (rej SP) are presented in Table 1. In skim milk, CN/TKN and CN/TP ratios are 0.77 – 0.79 and 0.8 – 0.83, respectively. The CN/TKN ratio ranged from 0.87 to 0.96, the highest ratio of 0.96 was obtained with 34.5 kPa pressure and 150% DF. The CN/TP ratio ranged from 0.89 to 0.96, the highest ratio of 0.96 was obtained with the use of 34.5 kPa TMP and 150% DF. For this combination, the highest SP removal was obtained. From the data, rej SP increased with operating pressure and decreased with the level of DF. The lowest rej SP of 0.1 was obtained with 34.5 kPa TMP and 150% DF combination. For all the runs, rej CN ranged from 0.97 to 1.0 by the membrane. Casein content of permeates ranged from 0.01 to 0.04%.

Table 1. Mean (n=3) data on casein to total protein ratio (CN/TKN), casein to true protein ratio (CN/TP), rejection of casein (Rej CN) and of serum protein (Rej SP) obtained from the lab-scale experiments

| DF (%) | 0 | 70 | 100 | 150 |
|--------|---|----|-----|-----|
| TMP (kPa) | 34.5 | 62.1 | 103.4 | 34.5 | 62.1 | 103.4 | 34.5 | 62.1 | 103.4 |
| CN/TKN | 0.89<sup>a</sup> | 0.89<sup>s</sup> | 0.87<sup>b</sup> | 0.94<sup>c</sup> | 0.93<sup>d</sup> | 0.9<sup>e</sup> | 0.94<sup>b</sup> | 0.93<sup>cde</sup> | 0.92<sup>o</sup> | 0.96<sup>a</sup> | 0.95<sup>b</sup> | 0.93<sup>cde</sup> |
| CN/TP | 0.90<sup>e</sup> | 0.90<sup>f</sup> | 0.89<sup>e</sup> | 0.94<sup>abc</sup> | 0.94<sup>c</sup> | 0.92<sup>e</sup> | 0.95<sup>b</sup> | 0.93<sup>cd</sup> | 0.96<sup>e</sup> | 0.95<sup>b</sup> | 0.93<sup>cd</sup> |
| Rej CN | 1.00<sup>a</sup> | 0.98<sup>abc</sup> | 0.99<sup>ab</sup> | 0.98<sup>abc</sup> | 0.99<sup>ab</sup> | 1.00<sup>a</sup> | 0.98<sup>abc</sup> | 0.97<sup>bc</sup> | 0.98<sup>abc</sup> | 0.96<sup>c</sup> | 0.97<sup>bc</sup> |
| Rej SP | 0.33<sup>c</sup> | 0.40<sup>b</sup> | 0.53<sup>d</sup> | 0.14<sup>c</sup> | 0.17<sup>c</sup> | 0.32<sup>cd</sup> | 0.13<sup>d</sup> | 0.17<sup>c</sup> | 0.27<sup>cd</sup> | 0.10<sup>e</sup> | 0.16<sup>df</sup> | 0.25<sup>d</sup> |

<sup>1</sup> DF = diafiltration; <sup>2</sup> TMP = transmembrane pressure.
<sup>a-b</sup> Mean values within same row not sharing a common superscript are significantly different (P < 0.05).

3.4. Pilot-scale experiments:

Pilot-scale experiments were conducted with 151.4 L of fresh skim milk for each experiment (Figure 6). A batch type pilot MF unit fitted with two SW (3830) PVDF membrane elements was used for MF of skim milk. Five pilot experiments were conducted at 34.5 kPa baseline and 103.4 kPa differential pressures, resulting in a TMP of 86.2 kPa (12.5 psi). These five runs used 100% DF. Two experiments were conducted at the same pressures, but with 150% DF. Experiments were also conducted at 62.1 kPa baseline and 103.4 kPa differential pressures, using 70 and 100% DF. All the experiments were conducted in continuous concentration mode, concentrating the feed to a final volume of 37.85 L giving a VR of 4. All the experiments were conducted at 24 °C. Deionized water was used for DF purposes.
Figure 6. The spiral-wound microfiltration (SW-MF) system used during the microfiltration of skim milk at different pressures and different diafiltration (DF) levels.

Using the model developed for lab-scale experiments, SP removal for pilot runs operated at above mentioned operating conditions was predicted. Actual SP removal data obtained during the experiments were compared with the predicted values. From the data presented in Table 2, the model predicted SP removal within 90-95% of the actual values.

Table 2. Comparison of SP removal predicted by the model and actual SP removal data obtained from the experiments

| Run # | Operating pressure, kPa | DF1, % | Serum protein removal, % |
|-------|------------------------|--------|--------------------------|
|       | Base | Differential | TMP2 | Actual | Predicted | Difference |
| 1     | 34.5 | 103.4        | 86.2 | 66.54  | 65.56     | -0.98      |
| 2     | 34.5 | 103.4        | 86.2 | 68.20  | 65.56     | -2.64      |
| 3     | 34.5 | 103.4        | 86.2 | 65.46  | 65.56     | 0.10       |
| 4     | 34.5 | 103.4        | 86.2 | 63.40  | 65.56     | 2.16       |
| 5     | 34.5 | 103.4        | 86.2 | 63.15  | 65.56     | 2.41       |
| 6     | 34.5 | 103.4        | 86.2 | 67.86  | 74.56     | 6.70       |
| 7     | 34.5 | 103.4        | 86.2 | 68.84  | 74.56     | 5.72       |
| 8     | 62.1 | 103.4        | 113.8| 53.80  | 51.89     | -1.91      |
| 9     | 62.1 | 103.4        | 113.8| 52.70  | 51.89     | -0.81      |
| 10    | 62.1 | 103.4        | 113.8| 61.33  | 57.29     | -4.04      |
3.5. Pilot Plant Process Flux

In MF of milk, for selective fractionation of milk components, it is essential to maintain the flux rates at reasonably close to initial flux rates. Any accumulation of polarization layer on the membrane surface and consequent drop in flux rates is detrimental to the fractionation process. Instantaneous flux rates obtained during pilot-scale runs for 86.2 kPa TMP at 100 and 150% DF and for 100% DF at 86.2 kPa and 113.8 kPa at 100% DF are presented in Figure 7.

Figure 7. Instantaneous flux obtained for 86.2 kPa TMP at 100 and 150% DF and for 113.8 kPa and 100% DF. The data are from the pilot plant run.

In these runs, DF water was added at VR of 1.23, 1.45, 1.68, 1.88, 1.88 and 2.67. After the final addition at a VR of 2.67, the process was continued to concentrate MCC till VR of 4. The valleys and peaks in the flux graphs are due to the addition of DF water at different intervals. The controlled addition of DF water could maintain the flux at or close to initial flux for 80% of the total process run time. In these pilot runs, the instantaneous flux was maintained within 90% of initial flux for about 80% of the process run. In the case of lab-scale experiments in which no DF was used, instantaneous flux dropped to about 60% of the initial value (Figure 2) with a consequent drop in flux and impairment of selective removal of SP. Addition of DF water in small quantities over several intervals of process run sustained the process flux rate for a longer time during the process and to maintain membrane selectivity.

3.6. Rejection of SP and CN - Pilot runs

The data on CN/TKN and CN/TP ratios and rejection of CN and of SP are presented in Table 3. From the data, 86.2 kPa TMP has a higher SP removal rate, higher CN/TKN, and CN/TP ratios and lower rejection of SP. MF process is pressure sensitive process. The results show a definite advantage of operating at lower TMPs. Results also show the effect of level of DF on SP removal and other parameters, higher DF levels showing definite advantage.
Table 3. Mean±SD of Serum protein removal (SP R, %), Casein to total protein (CN/TKN), Casein to true protein (CN/TP) ratio, and rejection of Casein (Rej CN) and serum protein (Rej SP) data obtained from pilot plant experiments

| Pressure | 34.5 base / 86.5 kPa TMP\(^2\) | 62.1 base / 113.8 kPa TMP\(^2\) |
|----------|-------------------------------|-------------------------------|
| DF\(^1\), % | Mean±SD (n=5) | Mean±SD (n=3) | Mean±SD (n=3) | - |
| SP R, % | 65.4±2.11 | 68.4±0.64 | 53.3±0.78 | 61.3 |
| CN/TKN | 0.92±0.01 | 0.92±0.00 | 0.90±0.01 | 0.91 |
| CN/TP | 0.92±0.01 | 0.93±0.01 | 0.90±0.00 | 0.92 |
| Rej CN | 0.98±0.00 | 0.99±0.01 | 0.99±0.00 | 0.98 |
| Rej SP | 0.27±0.02 | 0.27±0.01 | 0.38±0.01 | 0.30 |

\(^1\)DF= diafiltration; \(^2\)TMP= transmembrane pressure.

4. Conclusions

From the above presented results, the use of DF positively affected process flux rates and SP removal rate. Of various DF levels used in the study, 150% DF resulted in the highest impact on MF of skim milk. Over the range of operating pressures used in the study, the use of lower pressures resulted in higher SP removal and process flux rates. The results from this study show that use of lower pressures and controlled degrees of DF (addition of DF water at several intervals) will give maximized SP removal and process flux rates.

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