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# Effect of microfiltration concentration factor on serum protein removal from skim milk using spiral-wound polymeric membranes[1]

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## ABSTRACT

Our objective was to determine the effect of concentration factor (CF) on the removal of serum protein (SP) from skim milk during microfiltration (MF) at 50°C using a 0.3-μm-pore-size spiral-wound (SW) polymeric polyvinylidene fluoride (PVDF) membrane. Pasteurized (72°C for 16 s) skim milk was MF (50°C) at 3 CF (1.50, 2.25, and 3.00×), each on a separate day of processing starting with skim milk. Two phases of MF were used at each CF, with an initial startup-stabilization phase (40 min in full recycle mode) to achieve the desired CF, followed by a steady-state phase (90-min feed-and-bleed with recycle) where data was collected. The experiment was replicated 3 times, and SP removal from skim milk was quantified at each CF. System pressures, flow rates, CF, and fluxes were monitored during the 90-min run. Permeate flux increased (12.8, 15.3, and 19.0 $kg/m^2$ per hour) with decreasing CF from 3.00 to 1.50×, whereas fouled water flux did not differ among CF, indicating that the effect of membrane fouling on hydraulic resistance of the membrane was similar at all CF. However, the CF used when microfiltering skim milk (50°C) with a 0.3-μm polymeric SW PVDF membrane did affect the percentage of SP removed. As CF increased from 1.50 to 3.00×, the percentage of SP removed from skim milk increased from 10.56 to 35.57%, in a single stage bleed-and-feed MF system. Percentage SP removal from skim milk was lower than the theoretical value. Rejection of SP during MF of skim milk with SW PVDF membranes was caused by fouling of the membrane, not by the membrane itself and differences in the foulant characteristic among CF influenced SP rejection more than it influenced hydraulic resistance. We hypothesize that differences in the conditions near the surface of the membrane and within the pores during the first few minutes of processing, when casein micelles pass through the membrane, influenced the rejection of SP because more pore size narrowing and plugging occurred at low CF than at high CF due to a slower rate of gel layer formation at low CF. It is possible that percentage removal of SP from skim milk at 50°C could be improved by optimization of the membrane pore size, feed solution composition and concentration, and controlling the rate of formation of the concentration polarization-derived gel layer at the surface of the membrane during the first few minutes of processing.
**Key words:** microfiltration, concentration factor, serum protein, spiral-wound membrane

## INTRODUCTION

Cross-flow microfiltration (**MF**) of skim milk is a pressure-driven membrane separation that can be used to separate serum proteins (**SP**) from CN proteins in skim milk. Pore sizes of membranes used for MF range from 0.1 to 5.0 μm, depending on the application (Cheryan, 1998). Casein micelles (0.02 to 0.40 μm in diameter) are in colloidal suspension in milk, and are approximately 100 times larger than SP (0.003 to 0.010 μm), which are soluble in milk (Walstra et al., 2006). Retentates produced during MF of skim milk using a membrane with a pore size of 0.1 to 0.3 μm contain higher concentrations of CN micelles and other suspended particles, which are larger than the maximum membrane pore size. The CN-rich retentates, often called micellar casein concentrates (**MCC**), can be used in myriad of products including cheese (Nelson and Barbano, 2005), or other food and beverage products.

Membrane materials commonly used to microfilter skim milk to create MCC include ceramic (i.e., inorganic) and polymeric organic materials. Ceramic MF membranes used for the production of MCC have been studied (Zulewska et al., 2009; Hurt et al., 2010), and the advantages of their use compared with polymeric MF membranes include longer life and greater resistance to cleaning chemicals and higher temperatures (Cheryan, 1998). The most common configuration of polymeric membranes used by the dairy industry in

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North America is spiral-wound (**SW**) but other configurations (e.g., tubular, hollow fiber, and flat sheet) are possible. Spiral-wound polymeric MF membranes have the advantage of being less expensive to purchase (Cheryan, 1998), and SW membrane technology is familiar to the dairy industry, as it has been used since the 1970s for UF of cheese whey (Maubois, 1980). Most published research on MCC manufacture using MF has focused on ceramic membranes (Saboya and Maubois, 2000; Zulewska et al., 2009; Hurt et al., 2010) because of their recognized ability to separate SP from CN in skim milk. Ceramic membranes have a narrower pore size distribution than polymeric membranes (Brans et al., 2004), which gives ceramic membranes an enhanced ability to retain particles with diameters greater than a given pore size.

Currently, no commercial sales or utilization of MCC and serum protein concentrate exist as functional protein ingredients in foods. Experimentation with the manufacture and characterization of the functional properties of these products and comparison with similar chemically precipitated casein products or whey protein products will establish any economic or functional advantages of these dairy protein ingredients. Beliciu et al. (2012) and Sauer and Moraru (2012) had demonstrated that MCC may have advantages in retorted shelf-stable high-protein nutritional beverages. Opportunities may exist to use serum protein concentrate for protein beverage fortification due to their better clarity (Evans et al., 2009, 2010) and cleaner flavor profile than whey protein concentrates (Evans et al., 2009, 2010; Jervis et al., 2012; Campbell et al., 2013) for possible use in clear, low-pH, shelf-stable beverages. Clearly, the cost of ceramic membranes and the energy use are both high and have been concerns, making dairy processors cautious about investing in this technology even when their flux of $\geq$54 kg/m$^2$ per hour is attractive (Hurt et al., 2010). The cost of polymer MF membrane systems is much lower than ceramic systems (Cheryan, 1998), but their propensity to foul and exhibit low flux compared with ceramic MF membranes (Zulewska et al., 2009) is a concern and reduces their cost advantage. Low permeate flux (6 to 17 kg/m$^2$ per hour) during MF (with SW membranes) of skim milk have been reported (Lawrence et al., 2008; Zulewska et al., 2009; Beckman et al., 2010). Fouling of ceramic membranes during skim milk MF is minimized by high cross-flow velocities (e.g., 5 to 7 m/s) and a uniform transmembrane pressure (**TMP**; Saboya and Maubois, 2000). Not all configurations of ceramic membranes produce the same percent SP removal, and a recent study of Isoflux ceramic membranes (Technologies Avancees et Membranes Industrielles Nyons, France) reported a substantially lower (ca. 70%) SP removal for a 3-stage, 3.00× concentration process at 50°C (Adams and Barbano, 2013). Spiral-wound MF membranes operate at lower (e.g., <2 m/s) cross-flow velocities (Lawrence et al., 2008) and typically do not operate with a uniform TMP (Zulewska et al., 2009; Beckman et al., 2010), leading to much lower flux than that of ceramic membranes.

One of the largest disadvantages of SW polymeric MF membranes is the low efficiency of SP removal from skim milk. Zulewska et al. (2009) reported single-stage SP removal rates from skim milk at 50°C at 3.00× concentration factor (**CF**) of 64, 61, and 39% for ceramic uniform-TMP (**UTP**), ceramic graded-permeability, and SW polymeric membranes, respectively. For an SW MF system, Beckman et al. (2010) reported about a 70% SP removal from skim milk in a 3-stage 3.00× concentration process at 50°C compared with an SP removal of about 98% with UTP (Hurt et al., 2010) and 97% with graded-permeability ceramic membranes (J. Zulewska, Faculty of Food Science, University of Warmia and Mazury, Olsztyn, Poland, personal communication). In the report by Beckman et al. (2010), it was observed that flux increased with decreasing CF and we hypothesize that SP removal with SW membranes may be higher when the SW system is operated at lower CF, higher flux, and with less concentration polarization fouling.

Increasing the SP removal performance of SW polymeric MF would improve the chances of widespread implementation of this technology in the dairy industry and potentially increase the utilization of milk proteins in foods. An improved understanding of fouling of SW MF membranes and development of methods to improve polymeric membrane performance for separation of CN and SP from skim milk are needed. Our objective was to determine the effect of different MF CF (1.50, 2.25, and 3.00×) on SP removal from skim milk, and fouling of polymeric polyvinylidene fluoride (**PVDF**) SW 0.3-μm MF membranes at 50°C.

## MATERIALS AND METHODS

### *Experimental Design and Statistical Analysis*

On d 1 of processing, raw whole milk was centrifugally (4°C; model 372 Airtight; DeLaval Separator Co., Poughkeepsie, NY) separated at 5,000 × *g* into skim milk and cream. The skim milk was pasteurized (72°C for 16 s) and stored at 4°C. On d 2, 3, and 4, the pasteurized skim milk was MF at 50°C using a 0.3-μm-nominal-pore-size SW polymeric PVDF membrane, using 1 of 3 CF (1.50, 2.25, or 3.00×) on each of the days. On each day of filtration, 2 phases of the MF processing were run: startup-stabilization (about