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Micellar casein concentrate production with a 3X, 3-stage, uniform transmembrane pressure ceramic membrane process at 50°C[1]

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ABSTRACT

The production of serum protein (SP) and micellar casein from skim milk can be accomplished using microfiltration (MF). Potential commercial applications exist for both SP and micellar casein. Our research objective was to determine the total SP removal and SP removal for each stage, and the composition of retentates and permeates, for a 3×, continuous bleed-and-feed, 3-stage, uniform transmembrane pressure (UTP) system with 0.1-μm ceramic membranes, when processing pasteurized skim milk at 50°C with 2 stages of water diafiltration. For each of 4 replicates, about 1,100 kg of skim milk was pasteurized (72°C, 16 s) and processed at 3× through the UTP MF system. Retentate from stage 1 was cooled to <4°C and stored until the next processing day, when it was diluted with reverse osmosis water back to a 1× concentration and again processed through the MF system (stage 2) to a 3× concentration. The retentate from stage 2 was stored at <4°C, and, on the next processing day, was diluted with reverse osmosis water back to a 1× concentration, before running through the MF system at 3× for a total of 3 stages. The retentate and permeate from each stage were analyzed for total nitrogen, noncasein nitrogen, and nonprotein nitrogen using Kjeldahl methods; sodium dodecyl sulfate-PAGE analysis was also performed on the retentates from each stage. Theoretically, a 3-stage, 3× MF process could remove 97% of the SP from skim milk, with a cumulative SP removal of 68 and 90% after the first and second stages, respectively. The cumulative SP removal using a 3-stage, 3× MF process with a UTP system with 0.01-μm ceramic membranes in this experiment was 64.8 ± 0.8, 87.8 ± 1.6, and 98.3 ± 2.3% for the first, second, and third stages, respectively, when calculated using the mass of SP removed in the permeate of each stage. Various methods of calculation of SP removal were evaluated. Given the analytical limitations in the various methods for measuring SP removal, calculation of SP removal based on the mass of SP in the skim milk (determined by Kjeldahl) and the mass SP present in all of the permeate produced by the process (determined by Kjeldahl) provided the best estimate of SP removal for an MF process.

Key words: microfiltration, flux, serum protein, protein fractionation

INTRODUCTION

Casein micelles and serum proteins (**SP**) in skim milk can be separated by microfiltration (**MF**). This separation is possible because of the approximately 10- to 100-fold difference in diameter between CN micelles and SP (Walstra et al., 1999). A limitation of MF is membrane fouling, which reduces flux and can decrease transmission of SP (Sachdeva and Buchheim, 1997). One technique to minimize fouling is the use of cross-flow filtration, in which the retentate is pumped tangentially across the surface of the membrane. Increased flux seen in cross-flow filtration can be explained by the decrease in concentration polarization layer and lifting of solute particles away from the membrane surface due to shear at the membrane surface (Belfort et al., 1994). Le Berre and Daufin (1996) characterized the relationship between flux and shear rate at the membrane surface during MF of skim milk to separate CN micelles from SP. They found a critical ratio of flux to shear stress of 1.0 L/h per m^2 per Pa, where the pressure is the pressure decrease from the inlet to outlet of the membrane when operating a uniform transmembrane pressure (**UTP**) system at a concentration factor (**CF**) of 2× at 50°C, and that operating above this ratio led to decreased SP transmission and increased fouling.

In a standard cross-flow MF module a pressure drop will occur along the length of the membrane on the retentate side in the direction of fluid flow; in contrast, pressure on the permeate side of the membrane is relatively constant along the length of the membrane. This means the transmembrane pressure (**TMP**) and

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thus flux at the membrane inlet is higher than at the outlet and varies along the length of the membrane. No matter what operating conditions are chosen, parts of the membrane could be operating under nonideal conditions, leading to excessive fouling congruent with the concept of a critical flux to shear ratio. A solution to this problem was developed by Sandblom (1978), in which the permeate was recirculated on the permeate side of the membrane in the same direction as the retentate flow. The recirculation of permeate creates a pressure decrease on the permeate side of the membrane from inlet to outlet mirroring the pressure drop on the retentate side from inlet to outlet, creating UTP along the membrane's length (and uniform flux).

A UTP system requires membranes that are rigid and self-supporting, because they must be able to handle back pressure. This requirement rules out the use of most polymeric membranes, including spiral-wound membranes, in the UTP approach (Cheryan, 1998). Tubular ceramic membranes have been used successfully in UTP systems to separate CN micelles from SP (Nelson and Barbano, 2005; Zulewska et al., 2009). Some of the earliest published work was done by Pierre et al. (1992), using 0.2-μm ceramic membranes and concentrating skim milk to 3× before diafiltering. They found that removal of serum proteins from skim milk approached theoretical values. Le Berre and Daufin (1996) found that under optimal operating conditions, transmission of SP was 70 to 80%, and >99% of the CN was retained using a 0.1-μm ceramic UTP system where skim milk was concentrated to 2× at 50°C. Nelson and Barbano (2005) used a 3-stage, 3× UTP MF process with 0.1-μm ceramic membranes, with dilution using UF permeate between stages. They found an overall SP removal of 95% after 3 stages.

Both the micellar CN concentrate and SP separated by MF have the potential to be valuable products. The SP has been further purified by ultrafiltration to produce SP concentrates. Serum protein concentrates lack the glycomacropeptides present in whey protein concentrates and have a lower concentration of lipids (Evans et al., 2009). Serum protein isolates exhibit better foaming and gelling properties compared with whey protein concentrates (Britten and Pouliot, 1996). In addition, whey protein concentrates have been found to have diacetyl flavors that SP concentrates lack (Evans et al., 2009).

The micellar CN concentrate could be used to increase cheese yields and revenue (Papadatos et al., 2003) or potentially in food ingredient applications where caseinates are currently used. A single-stage UTP MF process with a CF of 3 using 0.1-μm ceramic membranes can remove >60% of the SP from the micellar CN (Nelson and Barbano, 2005; Zulewska et al., 2009); however, there could be advantages to using multiple stages to remove a greater percentage of SP, soluble minerals, and lactose from the micellar CN concentrate. Casein micelles are very heat stable (Holt, 1992, p.133), whereas whey proteins are not as heat stable and begin denaturing at 70°C (de Wit and Klarenbeck, 1984). Lactose also undergoes thermal degradation including Maillard reactions with proteins that can lead to off-flavors and browning (Walstra et al., 1999).

Theoretically, 97% of the SP should be removed from skim milk in a 3-stage, 3× MF process, but the actual removal and yield of micellar CN concentrate can be influenced by several operational parameters (Hurt and Barbano, 2010). No published research has determined the actual amount of SP, relative to theoretical values, that can be removed in a 3-stage UTP MF process with water diafiltration between stages. Our objective was to determine the total SP removal and the SP removal for each stage for a 3× continuous bleed-and-feed, 3-stage UTP system with 0.1-μm ceramic membranes, when processing pasteurized skim milk at 50°C with 2 stages of water diafiltration.

MATERIALS AND METHODS

Experimental Design and Statistical Analysis

One lot of bovine milk (approximately 1,099 kg) was separated in the Cornell University dairy plant at 4°C using a model 590 Air Tight centrifuge (DeLaval Co., Chicago, IL). Raw skim milk was pasteurized with a plate heat exchanger with 3 sections: regeneration, heating, and cooling (model 080-S, AGC Engineering, Manassas, VA) at 72°C with a holding time of 16 s. Temperature was kept at a minimum for pasteurization to minimize denaturation of SP. The milk was cooled to 4°C and stored at ≤4°C until processing. On d 1, pasteurized skim milk was heated to 50°C with a plate heat exchanger (model A3, DeLaval Inc., Kansas, MO) and microfiltered using a pilot-scale ceramic UTP system in bleed-and-feed mode to continuously produce a 3× MF retentate and MF permeate at 50°C. The MF retentate was cooled to ≤4°C as it was collected and stored until the next processing day. On the second day, MF retentate from the first day was diluted back to a 1× concentration (2 kg of water for every 1 kg of retentate) with pasteurized reverse osmosis (**RO**) water, heated to 50°C, and diafiltered with the ceramic UTP MF system to produce a 3× retentate. On the third day, this diafiltration procedure was repeated to complete a 3-stage process. This process was replicated 4 times starting with different batches of raw milk.

Data were analyzed by ANOVA using the Proc GLM procedures of SAS (version 8.02 1999-2001, SAS Insti-