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Effect of soluble calcium and lactose on limiting flux and serum protein removal during skim milk microfiltration[1]

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ABSTRACT

The tendency of calcium to promote microfiltration (MF) membrane fouling is well documented, but the role of lactose has not been studied. Milk protein concentrate that is 85% protein on a dry basis (MPC85) contains less calcium and lactose than skim milk. Our objectives were to determine the effects of skim milk soluble calcium and lactose concentrations on the limiting fluxes (LF) and serum protein (SP) removal factors of 0.1-μm ceramic graded permeability membranes. The MF was fed with 3 different milks: skim milk, liquid MPC85 that had been standardized to the protein content of skim milk with reverse osmosis water (MPC), and liquid MPC85 that had been standardized to the protein and lactose contents of skim milk with reverse osmosis water and lactose monohydrate (MPC+L). Retentate and permeate were continuously recycled to the feed tank. The LF for each feed was determined by increasing flux once per hour from 55 $kg \cdot m^{-2} \cdot h^{-1}$ until flux did not increase with increasing transmembrane pressure. Temperature, pressure drop across the membrane length, and protein concentration in the retentate recirculation loop were maintained at 50°C, 220 kPa, and 8.77 ± 0.2%, respectively. Experiments were replicated 3 times and the Proc GLM procedure of SAS was used for statistical analysis. An increase in LF between skim milk (91 $kg \cdot m^{-2} \cdot h^{-1}$) and MPC+L (124 $kg \cdot m^{-2} \cdot h^{-1}$) was associated with a reduction in soluble calcium. The LF of MPC+L was lower than the LF of MPC (137 $kg \cdot m^{-2} \cdot h^{-1}$) due to the higher viscosity contributed by lactose. Permeates produced from the MPC and MPC+L contained more protein than the skim milk permeate due to the transfer of caseins from the micelles into the reduced-calcium sera of the MPC and MPC+L. A SP removal factor was calculated by dividing true protein in the permeate by SP in the permeate portion of the feed to describe the ease of SP passage through the membrane. No differences in SP removal factors were detected among the 3 feeds below the LF. As the fluxes approached the LF, SP removal factors decreased due to fouling. Feeding a MF system with MPC instead of skim milk will reduce the required membrane surface area, but the permeate protein composition will be slightly higher in casein content.

Key words: microfiltration, limiting flux, serum protein, lactose

INTRODUCTION

Microfiltration (**MF**) is a membrane process designed to remove particulate matter (0.1 to 10 μm) from fluids. The dairy industry uses MF for applications such as whey defatting, bacterial cell removal, and protein fractionation of serum proteins (**SP**) and CN (Karleskind et al., 1995; Elwell and Barbano, 2006; Hurt et al., 2010). In milk, most CN exist as colloidal micelles that are 0.15 μm in diameter, on average. Soluble SP are roughly 100 times smaller. Treating skim milk with a 0.1-μm MF process should concentrate CN micelles in the retentate and allow SP to pass through the membrane into the permeate. Accumulation of soil on a membrane, or fouling, can limit SP transmission, thus making this separation less efficient. Fouling can also suppress permeate flux and make cleaning more difficult. These effects increase the costs of a MF plant.

Flux and the extent of fouling are dependent on the driving force of the filtration process, transmembrane pressure (**TMP**). The nature of this dependence can be explained by the critical flux theory (Brans et al., 2004). Briefly, 3 regimes exist wherein the TMP is below, slightly above, and well above a critical pressure, respectively. In the first regime, flux is linearly dependent on TMP according to Darcy's law (Hurt et al., 2015a,b). In the second regime, a critical flux is exceeded, fouling proceeds more rapidly, and flux becomes almost independent of TMP. The apex of this regime is known as the limiting flux (**LF**), or the maximum stationary flux that can be achieved by increasing TMP (Bacchin et al., 2006). The LF is a result of fouling

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and is dependent on cross-flow velocity, temperature, membrane design, and the feed composition (Samuelsson et al., 1997b). If TMP is increased further, the process enters the third regime and flux begins to decline. Processors should strive to maintain their operations within the second regime to maximize the average flux (Brans et al., 2004).

Because the various solids of milk interact differently with the membrane, the composition of the feed will influence the degree of fouling. Proteins constitute a large proportion of the foulant layer in many dairy filtration processes. Jimenez-Lopez et al. (2008) confirmed the role of CN micelles in 0.1-μm ceramic MF fouling by microfiltering model fluids composed of micellar CN powder, which had been reconstituted in 3 liquids: water, UF permeate (water plus lactose plus soluble minerals), or MF permeate (water plus lactose plus soluble minerals plus SP). They determined that CN contributed to about 90% of the resistance of the fouling layer. Although SP were not as detrimental to flux decline, they contributed to the foulant layer as well. These findings were in agreement with those of Zulewska and Barbano (2013), who observed that feeding a polymeric MF system with CN-free skim milk instead of skim milk produced a higher average flux (80 vs. 17 $kg \cdot m^{-2} \cdot h^{-1}$) and higher SP removal (59 vs. 35% reduction) during a continuous 3× process.

Mineral fouling is considered to be another important cause of flux decline in MF (Vetier et al., 1988). The mineral deposits formed are usually devoid of Mg, Na, K, S, and Cl, but Ca phosphate is always present (Hanemaaijer et al., 1989). Calcium phosphate can precipitate and form scale deposits on and within the membrane. Tan et al. (2014) determined that minerals did not significantly contribute to the fouling structure when microfiltering skim milk at 6°C using ceramic membranes. However, increasing process temperature may increase mineral fouling, as Ca phosphate solubility decreases with increasing temperature (Pouliot et al., 1989). Vetier et al. (1988) noted a substantial increase in mineral fouling after increasing the MF temperature from 20 to 50°C during ceramic MF of whole milk. The temperature effect is less pronounced above 50°C. Hurt et al. (2015a) were able to maintain constant flux during a skim milk MF process at temperatures between 50 and 65°C without increasing the TMP. Divalent cations such as Ca^{2+} can also contribute to fouling by facilitating electrostatic protein-protein interactions between the negatively charged carboxyl groups of milk proteins (Rice et al., 2009). Even in the absence of CN and SP, Ca has been implicated in fouling ceramic MF membranes (Jimenez-Lopez et al., 2008), although contributions of lactose and other nonprotein soluble components have not been experimentally ruled out. Lactose is not thought to be an important foulant in dairy filtration processes (Marshall and Daufin, 1995; Rice et al., 2009). Its role in UF flux suppression is limited to its effect on viscosity (Rao et al., 1994). However, to the best of the authors' knowledge, the effects of lactose on skim milk ceramic MF fouling have not been systematically decoupled from those of other soluble nonprotein milk components.

One method to reduce ceramic membrane fouling due to Ca phosphate might be to feed the MF system with a milk protein concentrate instead of skim milk. A milk protein concentrate is the retentate produced by UF and diafiltration of skim milk and is named according to its percentage of protein on a dry basis (i.e., a milk protein concentrate that is 85% protein on a dry basis is a **MPC85**). Relative to skim milk, soluble nonprotein component concentrations (i.e., lactose and soluble minerals) in MPC85 are reduced and protein concentrations are increased. If the ultimate goal of a MF process is to produce a micellar CN concentrate (Hurt et al., 2010) with a low lactose content, feeding the system with a MPC85 will decrease the required MF membrane area (Hurt and Barbano, 2015) and may increase the LF by mitigating Ca-induced fouling.

The objectives of this work were to determine the changes in LF and SP removal associated with reductions in lactose and soluble calcium in skim milk during a 0.1-μm ceramic graded permeability (**GP**) membrane MF process at 50°C. Skim milk, MPC85, and MPC85 with added lactose were each microfiltered while flux was increased from 55 $kg \cdot m^{-2} \cdot h^{-1}$ to the LF. The compositions and viscosities of the feeds, retentates, and permeates from each process were determined and used to explain the results.

MATERIALS AND METHODS

Experimental Design

A completely randomized split block design was employed in which feed type was the whole plot factor, flux was the sub plot factor, and replicate was the blocking factor. The experiment was replicated 3 times in different weeks with 3 different batches of fresh pasteurized skim milk and liquid MPC85 (12% protein, 0.5% lactose, 0.2% fat, 14% total solids) made from that skim milk. Each replicate took place over 4 d in a week. On the first day, HTST-pasteurized (73°C, 20 s) skim milk (300 kg) and liquid MPC85 (200 kg) were procured from OATKA Milk Products Cooperative Inc. (Batavia, NY). The second, third, and fourth days were used for MF of the 3 feeds described below. The order of feed MF was randomized within each week and balanced among replicates.