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Separation of milk fat globules via microfiltration: Effect of diafiltration media and opportunities for stream valorization

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ABSTRACT

Milk fat globule membranes (MFGM) sourced in buttermilk have gained recent interest given their nutritional value and functional properties. However, production of isolated MFGM has been challenging given their size similarity with casein micelles, which limits attempts toward fractionation by size exclusion techniques. Therefore, the hypothesis underpinning this study is that the removal of proteins from cream before butter-making facilitates MFGM isolation. As such, milk fat globules were separated from raw whole milk via microfiltration (1.4 μm pore diameter and 0.005 m^2 filtration surface area) by using 3 diafiltration media; namely, skim milk ultrafiltration permeate, saline, and water. Their effects on the stability of the milk fat globules and protein permeation was elucidated. Whereas a substantial reduction in protein concentration was achieved with all diafiltration media (~90% reduction), water and saline produced negligible membrane fouling with better filtration performance. Moreover, diafiltration with skim milk ultrafiltration permeate exhibited reduced permeate flux. Colloidal stability of the resultant milk decreased with all diafiltration solutions due to changing composition and reduced apparent viscosity. Overall, microfiltration was found to be an efficient method for separation of milk fat globules from whole milk, leading to increased MFGM fragment concentration in buttermilk dry matter, thus making it more suitable for industrial utilization.
Key words: diafiltration, microfiltration, milk fat globule membrane, milk fat separation

INTRODUCTION

Cream churning in butter processing disrupts the membrane of milk fat globules, and the resulting membrane fragments, known as a source of functional ingredients, are released into the aqueous phase, buttermilk. Thus, recent attention surrounding buttermilk as a potential value-added ingredient in dairy products is not surprising (Corredig et al., 2003; Le et al., 2011; Phan et al., 2013, 2014). The advantageous nutritional and functional properties and the various isolation methods of milk fat globule membrane (**MFGM**) have been reviewed (Dewettinck et al., 2008; Holzmüller and Kulozik, 2016). Furthermore, many efforts have been applied to fractionate and concentrate MFGM fragments from buttermilk, for example, via filtration (Corredig et al., 2003; Rombaut et al., 2006; Fauquant et al., 2014). Such MFGM isolation is based on the selective removal of casein, whey proteins, lactose, and minerals, of which the similarity in size of casein micelles and MFGM fragments has been reported to be the major obstacle for fractionation (Dewettinck et al., 2008). Traditionally milk is separated into cream and skim milk by mechanical separation based on differences in colloidal particle velocity under a centrifugal field. Such separation leads to the enrichment of low-density particles, fat globules, into cream, but it is not selective to proteins. From cream, proteins enter into the butter-making process and eventually end up in buttermilk. Unlike centrifugal separation, membrane filtration is a pressure-driven process that fractionates colloidal particles from the suspension by size exclusion. Membrane filtration enables protein separation from cream before butter-making for more efficient raw material usage. Hence, if most proteins are transferred to the skim milk, MFGM fragments can be fully exploited from buttermilk with minimal protein interference.

Milk is a complex system that can be fractionated, for example by cross-flow microfiltration (**MF**) with diafiltration (**DF**), which critically affects its physicochemical characteristics, such as a colloid diameter, electrostatic charge, hydration, mineral and protein composition, and so on (Jimenez-Lopez et al., 2011). The properties of milk play important roles not only in terms of milk functionality, but also in the effectiveness of related filtration processes. It is important to understand the interactions between milk components

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to control milk integrity and filtration performance. As a complex layer of surface material, MFGM naturally emulsifies the fat globules in milk. The structure of MFGM has been extensively studied and reported to consist of a phospholipid trilayer and a complex mixture of proteins and cholesterol (Fong et al., 2007; Gallier et al., 2012; Zou et al., 2015). The outer layer of the MFGM consists mainly of protruding glycocalyx and peripheral proteins (Singh, 2006), along with related structures that are considered native milk fat globules. Processing of the fat globules can lead to their coalescence and loss of membrane material, which enables the incorporation of proteins to resurface the fat globules and thus alters their physical properties. Washing cream can also selectively remove MFGM components from the interface and create accessible surfaces for casein and whey protein attachment (Kathriarachchi et al., 2014). In milk, caseins are present in the form of colloidal particles, the so-called casein micelles, which contain some inorganic matter, mainly calcium phosphate (Walstra et al., 2006). Caseins tend to associate due to hydrophobic effects and have a fairly high charge caused by the phosphate groups, which strongly binds divalent ions such as Ca^{2+}. Casein micelles undergo ion exchange with free ions; for example, addition of NaCl induces the displacement of casein-bound protons (Huppertz and Fox, 2006). Dynamic equilibrium of the casein micelle is primarily dependent on concentration, temperature, pH, and ionic strength (as well as other physicochemical conditions). To enhance filtration performance, DF is commonly used in dairy processing, which mainly involves washing out permeating compounds with DF medium. Use of DF alters the ionic environment surrounding the casein micelles and, by disruption of the micelles, lead to losses of colloidal calcium phosphate and casein proteins into the serum phase (Ferrer et al., 2014). The reduction of pH, or the addition of citrate or salt to milk, decreases the net surface charge of casein micelles and induces partial dissolution of the colloidal calcium that favors micelle disintegration (Jimenez-Lopez et al., 2011).

Milk membrane filtration systems are being exploited by the dairy industry for fractionation of milk proteins and removal of microorganisms (i.e., cold pasteurization; Beolchini, 2005; Hoffmann et al., 2006; Hurt and Barbano, 2010; Piry et al., 2012; Adams and Barbano, 2013). Skim milk is often used for milk membrane filtration processes, and filtration of fat-containing dairy fluids is mainly limited to defatting of whey (Golbandi et al. 2013). However, membrane filtration has been applied to separation of milk fat populations into small and large globules by MF (Goudedranche et al., 2000; Michalski et al., 2006) and for separation of anhydrous milk fat into fractions with different melting points (Abbas et al., 2006). Microfiltration of milk fat-rich dairy fluids is limited at the industrial scale, as milk fat has a role in membrane fouling. Fouling is a major challenge in all applications related to membrane filtration of milk. Fouling and concentration polarization are related to the deposition of a layer on the membrane surface and to the narrowing of the pores, which negatively affect the selectivity and filtration performance by flux reduction (Guerra et al., 1997; Fouladitajar et al., 2015). Concentration polarization, the formation of a dense, multicomponent layer at the walls of the filtration membrane, has remained one of the most important problems in pressure-driven membrane processes (Tashvigh et al., 2015). One way to prevent particle deposition and minimize fouling is to facilitate relatively higher shear at the membrane-liquid interface, which can be accomplished by use of high cross-flow velocities (4–8 m/s; Guerra et al., 1997). In addition to shear stress, MF performance can be controlled by the transmembrane pressure (**TMP**) and permeate flux (Gésan-Guiziou et al., 1999). High feed concentration and pressure leads to the formation of a thicker concentration polarization layer on the membrane wall (Fouladitajar et al., 2015). The formation of such a fouling layer can be inhibited by lowering the pressure and feed concentration and, in the case of whole milk MF, by reducing feed viscosity (e.g., by operating at temperatures above 50°C).

The MF of fat-containing dairy fluids can enable the separation of specific milk fat components, including MFGM fragments. Because of interactions between MFGM components during their isolation and the size similarity with casein micelles, the full potential of MFGM remains an untapped opportunity. Morin et al. (2007) investigated the possibility of removing caseins from cream by centrifugal separation before buttermaking. It was found that by washing cream with skim milk ultrafiltration permeate the protein content in the cream decreased by 60%, resulting in more effective MFGM fragment concentration from buttermilk. However, washing cream has been reported to release MFGM material and increase fat globule coalescence (Holzmüller et al., 2016). In addition, possible advantages of membrane separation over centrifugation include reduced energy consumption and less damage to shear-sensitive components, such as the MFGM (Brans et al., 2004). Furthermore, MF has not yet been considered for casein and milk fat separation. To the best of our knowledge, no reports exist on the application of MF toward native milk fat globule separation from milk or cream. Moreover, despite the extensive research devoted to the effect of minerals on the properties of milk proteins during filtration (Mao et al., 2012; Carpintero-Tepole et al., 2014; Ferrer et al., 2014), their