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The effect of butter grains on physical properties of butter-like emulsions

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ABSTRACT

Milk fat exists as globules in its natural state in milk. The potential of using globular fat to modulate the rheological properties and crystallization behavior in butter-like emulsions was studied in the present work. We conducted a comparative study of butter-like emulsions, with a fat phase consisting of 0, 10, 25, 50, or 100% anhydrous milk fat (AMF), the remaining fat being butter grains, and all samples containing 20% water, to obtain systematic variation in the ratio of globular fat. All emulsions were studied over 4 wk of storage at 5°C. By combining small and large deformation rheology, we conducted a detailed characterization of the rheological behavior of butter-like emulsions. We applied differential scanning calorimetry to monitor thermal behavior, confocal laser scanning microscopy for microstructural analysis, and low-field pulsed nuclear magnetic resonance spectrometry to measure solid fat content. By combining these techniques, we determined that increasing the fraction of globular fat (by mixing with butter grains) decreases the hardness of butter-like emulsions up to an order of magnitude at d 1. However, no difference was observed in thermal behavior as a function of butter grain content, as all emulsions containing butter grains revealed 2 endothermal peaks corresponding to the high (32.7°C ± 0.6) and medium (14.6°C ± 0.1) melting fractions of fatty acids. In terms of microstructure, decreasing the amount of butter grains in the emulsions resulted in formation of a denser fat crystal network, corresponding to increased hardness. Moreover, microstructural analysis revealed that the presence of butter grains resulted in faster formation of a continuous fat crystal network compared with the 100% AMF sample, which was dominated by crystal clusters surrounded by liquid oil. During storage, hardness remained stable and no changes in thermal behavior were observed, despite an increase in solid fat content of up to 5%. After 28 d of storage, we observed no difference in either microstructural or rheological properties, indicating that formation of primary bonds occurs primarily within the first day of storage. The rheological behavior of butter-like emulsions is not determined solely by hardness, but also by stiffness related to secondary bonds within the fat crystal network. The complex rheological behavior of milk fat-based emulsions is better characterized using multiple parameters.

Key words: milk fat, material science, butter grains, rheology

INTRODUCTION

Milk fat exists in its natural state as fat globules and is mainly composed of a triglyceride (**TG**) core surrounded by a complex protein membrane: the milk fat globule membrane. Anhydrous milk fat (**AMF**), consisting primarily of TG, is widely used in the food industry. Milk fat is characterized by an extreme diversity of FA, and thereby diverse TG composition, which induces a broad thermal range of melting transitions, ranging from −40°C to 40°C. The melting profile of milk fat is typically divided into 3 characteristic melting fractions: the low melting fraction with a melting point below 10°C, the medium melting fraction, which melts from 10°C to 19°C, and the high melting fraction, which melts above 20°C (Deffense, 1993). Despite the similar FA composition of nonemulsified bulk fat and emulsions such as natural cream (oil-in-water emulsion), their crystallization behavior differs (Walstra and Van Beresteyn, 1975; Fredrick et al., 2011). For fat-based systems, crystallization proceeds more slowly in oil-in-water emulsions compared with bulk fat because nucleation has to occur in each single globule. It has been argued that nucleation in intact and pure milk fat globules is presumably homogeneous, as the content of impurities is lower than in bulk fat and butter (Fredrick

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et al., 2011). Homogeneous nucleation requires more supercooling than heterogeneous nucleation, which therefore explains the slower crystallization process for emulsified fat. However, a previous study has reported a correlation between the size of milk fat globules and occurrence of heterogeneous nucleation behavior of TG (Michalski et al., 2004). Increasing the size of the milk fat globules increases the probability of having at least one catalytic impurity present within the milk fat globule, hence facilitating heterogeneous nucleation (Walstra, 2003). Moreover, the time needed to obtain the first nucleus catalyzing the crystallization process is inversely proportional to the size of the milk fat globules (Lopez et al., 2002, 2007). Another aspect to consider for butter-like emulsions is that the molecules located at the surface of the milk fat globules may induce crystallization of TG, conversely facilitating a faster crystallization process. Differences are reported in the complex and not yet fully understood crystallization mechanisms in bulk milk fat compared with emulsified fat (both oil-in-water and water-in-oil emulsions). However, the physical properties of the final crystallized products, as well as their final polymorphic crystal forms appear to be the same (Lopez et al., 2001a,b; Michalski et al., 2004; Rønholt et al., 2012).

The nucleation process and crystal growth are crucial for the fat crystal network, and consequently spreadability, mouthfeel, appearance, and product functionality (Mulder and Walstra, 1974). In addition, changing the amount of fat globules in milk fat-based products might influence product properties. Although of major industrial interest, the rheological response to the presence of fat globules within a continuous fat phase is documented in only a limited number of studies. It is generally accepted that the presence of fat globules decreases the hardness of the product compared with milk fat-based products without any fat globules present, because a crystalline intraglobular phase is formed rather than large bulk fat crystals (Mulder and Walstra, 1974; Juriaanse and Heertje, 1988; Buldo and Wiking, 2013). The microstructure of milk fat-based products (e.g., butter and butter blends) is very complex, due to a continuous fat crystal network interrupted by intact milk fat globules and water droplets, all dispersed in a continuous liquid fat phase (Juriaanse and Heertje, 1988; Buldo, 2013; Rønholt et al., 2013). When changing the ratio between the amounts of fat in the continuous phase versus fat within the fat globules, the microstructure changes accordingly and will likely affect the crystallization process, as discussed above; thus, microstructure formation and rheological properties within the system.

The fat crystal network is often characterized by the presence of irreversible strong bonds (e.g., as occurring due to mechanical interlinkage of the fat crystal upon crystal growth) and reversible weak bonds occurring via van der Waals interactions (van den Tempel, 1958). When applying a small stress, the crystal network may distort, resulting in reversible breakage of some crystal bonds. As the stress is removed, the crystal bonds will reform. In contrast, large stress will rearrange the network or cause fracturing, resulting in irreversible breaking of the bonds (Wright et al., 2001). The stress needed to break irreversible crystal-crystal interactions can be quantified by identification of the fracture point upon large deformation rheology, and the magnitude of this stress may in turn be linked to product hardness. The weak and reversible bonds are related to the elastic modulus of the material (Haighton, 1963). The rheological properties of milk fat are typically studied by using large or small deformation techniques. The rheological response to large deformations is highly nonlinear and involves breakdown of the material (Wright et al., 2001). In contrast, small deformations obtained within the linear region links material behavior to the applied deformation without breaking the structure of the product, characterizing the reversible, elastic bonds. As fat texture is a complex concept, it cannot be described with a single parameter (Mortensen and Danmark, 1982; Buldo, 2013). We therefore used both large deformation in the compression mode (stress at fracture) and small amplitude oscillation measurements (elastic modulus) using parallel plates to obtain a better rheological characterization of our emulsions.

The aim of this work was to elucidate how the presence of milk fat globules in a butter-like emulsion affects the crystallization process, microstructure formation, and thus rheological and textural properties. To do so, butter grains, containing both intact and partially damaged milk fat globules, were mixed in increasing amounts with AMF and water to form water-in-oil emulsions. The emulsions were stored at 5°C for 28 d and rheological and textural properties were monitored during storage. Furthermore, the crystallization behavior was studied using differential scanning calorimetry and linked to rheological properties as well as microstructure.

MATERIALS AND METHODS

Materials

Pasteurized cream with 38% fat (Arla Foods, Slagelse, Denmark) and AMF from Arla (Götene, Sweden) were used to prepare the emulsions. Sodium azide from Sigma Aldrich (St. Louis, MO) was added to the cream (0.2 g/L) in all emulsions to prevent microbial growth. For the confocal laser scanning microscopy (**CLSM**)