



Influence of homogenization on physical properties of model coffee creamers stabilized by quillaja saponin



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ABSTRACT

There is a growing demand for use of natural ingredients in food manufacturing. This study utilized a natural emulsifier, quillaja saponin (1%) to fabricate non-dairy model creamer emulsions (containing 10% medium chain triglycerides oil). Varying homogenization conditions, ranging from a high-shear mixer to passing through a microfluidizer at 20,000 psi, were applied to fabricate emulsions. The effect of particle size on the appearance, tristimulus color coordinates, and electrical characteristics of the model creamers and white coffee drinks were investigated. The average droplet size varied from 0.2 to 16 μm . All model creamers had whitish milk-like appearance and the white coffee solutions had light brown color. All systems were physically stable except for the systems with largest oil droplets (1.8 and 16 μm), which had creaming. The lightness, L^* (whiteness) of the model creamer and the white coffee increased with decreasing oil droplet size, as smaller droplets scatter more light. Decreasing the oil droplet size led to lower zeta potential (from -73 to -54 mV) due to lesser negative charge group accumulated on the interfacial layer of the droplets. The oil droplets were also found to be stable to aggregation in hot acidic coffee solutions prepared using model hard water. Overall, this study found that oil droplets stabilized with natural plant-based surfactant have potential for application in liquid coffee creamers and their stability and whitening power were dependent on the droplet size.

1. Introduction

Consumers are paying more attention to the ingredients used to formulate food products and many of them are making a deliberate effort to choose products that utilize natural, environmentally friendly, and sustainable ingredients (Euromonitor, 2016). As a result, food manufacturers are searching for natural microbial- or plant-based alternatives to many synthetic or animal-derived functional ingredients used in foods (McClements & Gumus, 2016). Synthetic ingredients (mono- and di-glycerides, fatty acid esters of mono- and di-glycerides) and animal-derived ingredients (such as milk, egg, or meat proteins) are commonly used as emulsifiers to facilitate the formation and stability of emulsion-based food and beverages, such as creamers, sauces, dressings, soups, desserts, infant formula, beverages, and ice cream (Dickinson, 2009; Petrut, Danthine, & Blecker, 2016). Consequently, there is a considerable research effort in identifying, isolating, purifying, and characterizing both microbial-based and plant-based emulsifiers suitable for utilization within food products (McClements & Gumus, 2016). To function as a suitable label-friendly emulsifier, a natural ingredient needs to exhibit certain

physicochemical and economic traits: (a) it should rapidly adsorb to the surfaces of the droplets formed during homogenization; (b) it should reduce the oil-water interfacial tension by an appreciable amount so as to facilitate further droplet disruption within the homogenizer; (c) it should generate strong repulsive forces between the droplets to stabilize them against aggregation during and after homogenization; (d) it should be easily dispersed in either the aqueous and/or the oil phases; (e) it should be readily available in sufficient quantities at a reliable quality and cost; and (f) it should be obtained from a sustainable natural resource (Dickinson & Leser, 2013; McClements, 2015).

Quillaja saponin, a natural small molecule surfactant, has been the subject of considerable research in recent years due to its excellent stabilizing, emulsifying, and foaming properties (Golemanov, Tcholakova, Denkov, Pelan, & Stoyanov, 2014; Toppel, Reim, Rohn, & Drusch, 2016; Yang, Leser, Sher, & McClements, 2013; Zhang & Reineccius, 2016). Quillaja saponins are extracted from the soapbark tree (*Quillaja saponaria*), which typically grows in certain regions of South America. Saponins are good emulsifiers due to their amphiphilic structure: they have a hydrophobic aglycone portion connected to a hydrophilic sugar portion (Golemanov et al., 2014;

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Orczyk & Wojciechowski, 2015; Stanimirova et al., 2011). Oil-in-water emulsions stabilized by quillaja saponin have been reported to be relatively stable over a wide pH range, *i.e.*, pH 3 to 7 (Ozturk, Argin, Ozilgen, & McClements, 2014; Yang et al., 2013). Studies have also reported that this natural surfactant can be used at relatively low surfactant-to-oil ratios (< 0.1) to form stable emulsions containing small droplets ($d < 200$ nm) (Ozturk et al., 2014; Yang et al., 2013). Consequently, quillaja saponins may be suitable for application in a wide range of different food and beverage products.

In this study, we used a quillaja saponin extract (Q-Naturale® 200, Ingredient) to formulate non-dairy model creamers designed for hot coffee applications, as most commercial coffee creamers are commonly made with milk proteins. Moreover, the National Coffee Association of the USA (2017) has reported that an increasing number of Americans drink coffee on a daily basis, with a rise from 57% to 62% from 2016 to 2017. In view of the rise, coffee creamers made from plant-based emulsifiers will provide more options for consumers who seek alternatives to animal-based ones. A good coffee creamer should have a number of characteristics: it should remain stable after addition to hot, acidic coffee solutions; it should whiten the coffee; it should neutralize the acidity; and it should impart desirable textural and mouthfeel properties (Golde & Schmidt, 2005; Khatkar & Gupta, 2014; Oldfield, Teehan, & Kelly, 2000). Oil-in-water emulsions used as creamers have to be carefully designed so that they do not destabilize during storage or after addition to coffee, which may occur through a number of mechanisms, including *feathering* (flakes on surface and/or throughout beverage), *creaming* (oil droplets on surface), *de-oiling* (free oil on surface), or *sedimentation* (sediments at bottom). Destabilization typically occurs due to aggregation of either the protein molecules and/or the oil droplets within creamers, which may be the result of a number of factors: *thermal aggregation* caused by the high temperatures in coffee; *isoelectric aggregation* caused by the fact the pH in white coffee may be near to the isoelectric point of free or adsorbed proteins; and *electrostatic aggregation* caused by screening of electrostatic repulsive interactions in the presence of soluble salts (Burgwald, 1923; Hamboyan, Pink, Klapstein, MacDonald, & Aboud, 1989; Tran & Einerson, 1987).

The ability of creamers to whiten coffees depends on the size and concentration of any colloidal particles they contain that scatter light, such as oil droplets or protein aggregates (McClements, 2002). In particular, the whitening power of a suspension of oil droplets is strongest when they have a droplet diameter around 200 nm (Keowmaneechai & McClements, 2002; Zhang & Reineccius, 2016). In the present study, the effect of oil droplet size on the physical properties of non-dairy creamers and white coffee was therefore investigated. Model creamers of varying particle sizes were fabricated using three mechanical devices, a high-shear mixer, a high-pressure valve homogenizer, and a microfluidizer. The visual appearance, tristimulus color coordinates, physical stability, and electrical characteristics of the model creamer and white coffee formed from it were then determined. The insights obtained from this study provide valuable knowledge about the application of a natural emulsifier (quillaja saponin) in commercial food products that have specific technical requirements, such as non-dairy creamers that must remain stable during ambient storage and after being added to hot acidic coffee solutions.

2. Materials and methods

2.1. Materials

Quillaja saponin (Q-Naturale 200®) was obtained from Ingredient Incorporated (Westchester, Illinois, USA). Medium chain triglyceride (Miglyol 812N) oil was purchased from Warner Graham Company (Cockeysville, MD, USA). A commercial dark roast coffee powder (Nestlé Nescafé Clásico) was purchased from a local supermarket. All analytical grade chemicals used, including hydrochloric acid, sodium hydroxide, sodium phosphate monobasic, sodium phosphate dibasic,

calcium chloride, and magnesium chloride were purchased from Fisher Scientific Company LLC (Pittsburgh, Pennsylvania, USA). All concentrations are presented on a percentage weight-to-weight basis (*i.e.*, % w/w) unless otherwise stated.

2.2. Methods

2.2.1. Fabrication of model creamer at varying pressure and passes

Model creamers containing 10% medium chain triglycerides (MCT) and 1% quillaja saponin were fabricated using a high-shear mixer, high-pressure valve homogenizer, or microfluidizer operated at different energy inputs, *i.e.*, rotational speed or homogenization pressure. A weighed amount of quillaja saponin solution was dispersed in 10 mM phosphate buffer (pH 7) to make a homogeneous aqueous phase. Coarse emulsions were then prepared by blending 10% oil phase with 90% aqueous phase using a high-shear mixer (Bamix, Biospec Products, Bartlesville, Oklahoma, USA) (15,000 rpm) for 1 min. One portion of the coarse emulsion was set aside for further use and the other portion was immediately passed once through either a high-pressure valve homogenizer operated at 1000 to 5000 psi (LAB 1000, APV-Gaulin, Wilmington, MA, USA) or a microfluidizer (Microfluidizer M-110P, Microfluidics, Newton, MA, USA) operated at 7500 to 20,000 psi. Some samples were passed through the microfluidizer multiple times (1 to 4) at constant pressure of 20,000 psi to obtain emulsions with different sizes. After fabrication, all emulsions were adjusted to pH 7 prior to analysis and addition to coffee solutions to mimic the pH conditions in commercial coffee creamers.

2.2.2. Addition of model creamer to black coffee

Initially, 1% black coffee solutions were prepared by reconstituting coffee powder in boiled hot hard water (270 ppm calcium carbonate, CaCO_3). The hard water was prepared by dissolving 1.95 mM calcium chloride and 0.62 mM magnesium chloride in Milli-Q water (18 $\text{M}\Omega\text{-cm}$ resistivity) to mimic mineral content of the hard water found in some local drinking waters. Immediately after making the hot black coffee, 10 mL of model creamer was added to 60 g of hot black coffee solution ($\sim 85^\circ\text{C}$) and stirred for 3 s. The mixture of hot coffee solution and model creamer is hereafter referred to as “white coffee”.

2.2.3. Characterization of model creamer and coffee systems

2.2.3.1. Particle size measurements. The particle size of the model creamer and coffee systems was measured using a laser diffraction particle size analyzer (Beckman Coulter LS 12 320, Brea, CA, USA). A few drops (0.1 to 0.5 mL) of samples were added into a measurement cell containing either 0.01 M phosphate buffer pH 7 for the model creamer measurements or hard water (270 ppm CaCO_3) for the coffee solution measurements to achieve an optimum obscuration rate of 40 to 55%. A refractive index of 1.333 was set for the aqueous phase and 1.448 for oil phase. The particle size distribution calculated by the instrument was based on finding the best fit between the predictions of the Mie theory and the measured light scattering pattern. The particle size measurements are reported as volume-weighted mean diameters, $d_{4,3}$.

2.2.3.2. Confocal laser scanning microscopy. The microstructure of the model creamer and coffee solutions was examined using confocal laser scanning microscope (Nikon D-Eclipse C1 80i, Nikon, Melville, NY, USA). Samples were prepared by mixing 1 mL of creamer or coffee with 0.1 to 0.2 mL of Nile red (1 mg Nile red/1 mL ethyl alcohol) to stain the oil phase, and then a small aliquot of the stained sample was placed onto a microscope slide and covered with a glass cover. The excitation and emission spectrum for the Nile red stain were 543 nm and 605 nm, respectively. The samples were examined with a $10\times$ eyepiece and $60\times$ objective lens and the microstructural images were captured and processed using image analysis software (EZ-CS1, Nikon, Melville, NY, USA).

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