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Production of highly concentrated oil-in-water emulsions using dual-channel microfluidization: Use of individual and mixed natural emulsifiers (saponin and lecithin)

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ABSTRACT

The fabrication of concentrated oil-in-water emulsions is useful for reducing storage and transportation costs, as well as for providing desirable textural, optical, stability, and release characteristics in commercial products. In this study, 50 wt% oil-in-water emulsions were produced from natural emulsifiers using high-pressure dual-channel microfluidization (89.6 MPa, 1 pass). The particle size and charge characteristics of emulsions stabilized using a hydrophilic biosurfactant (quillaja saponin) or mixtures of hydrophilic and hydrophobic biosurfactants (quillaja saponin + soy lecithin) were measured. The physical stability of the emulsions was determined during storage under quiescent conditions (pH 7, 25 °C). The mean droplet diameter and polydispersity decreased with increasing hydrophilic and hydrophobic biosurfactant concentration. Surface potential measurements indicated that interfacial composition depended on the amount of hydrophilic and hydrophobic biosurfactant present. The inclusion of hydrophobic emulsifier in the oil phase and hydrophilic emulsifier in the aqueous phase prior to homogenization, led to the formation of smaller oil droplets than using the hydrophilic emulsifier alone. The relatively small size and polydispersity of the droplets in the mixed-emulsifier systems led to a higher emulsion viscosity and a better aggregation stability, *i.e.*, there was a smaller change in particle size during storage. However, some creaming was still observed in the emulsions due to the presence of a fraction of relatively large droplets. In summary, concentrated emulsions stabilized by mixed biosurfactants may be advantageous for commercial application in certain food, beverage, and pharmaceutical products.

1. Introduction

Oil-in-water emulsions are typically fabricated in the food industry by homogenizing an oil and aqueous phase together in the presence of an emulsifier using mechanical devices, such as high shear mixers, colloid mills, high pressure valve homogenizers, microfluidizers, and sonicators (Friberg, Larsson, & Sjoblom, 2003; McClements, 2015; Schubert & Engel, 2004). The production of dilute emulsions containing fine droplets is often relatively straightforward, but for certain applications it is advantageous to produce highly concentrated emulsions (≥50%). The appearance, texture, flavor profile, and stability of emulsions is influenced by their droplet concentration, *e.g.*, emulsion viscosity, lightness, flavor retention, and creaming stability all tend to increase with increasing droplet level (McClements, 2015). Consequently, food manufacturers often need to produce concentrated emulsions so as to obtain the quality attributes expected in a particular product, such as the "creaminess" of creams, sauces, salad dressings, and mayonnaise. Studies have shown that the rate of lipid oxidation in oil-in-water emulsions is reduced when the total oil level is increased (Osborn & Akoh, 2004), and so increasing droplet concentration may improve the shelf-life of chemically labile products. The fabrication of highly concentrated emulsions may also be useful to reduce storage and transportation costs (Lissant, 1966; Piorkowski & McClements, 2014). In this case, a concentrated emulsion is initially created, which

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is then diluted when it is applied in the final product. Finally, concentrated emulsions may be used to increase the loading capacity of emulsion-based delivery systems for hydrophobic bioactives (Muller, Harden, & Keck, 2012). The ability to produce concentrated emulsions containing small droplets depends on homogenizer type and operating conditions, as well as the nature and concentration of emulsifiers used (Jafari, Assadpoor, He, & Bhandari, 2008; Walstra, 1993). In particular, the droplet collision frequency tends to increase with increasing droplet concentration, leading to a greater extent of droplet coalescence within the homogenizer (Gupta, Eral, Hatton, & Doyle, 2016; Jafari et al., 2008; Mohan & Narsimhan, 1997; Raikar, Bhatia, Malone, & Henson, 2009). Recent research in our laboratory has shown that stable concentrated emulsions can be produced by dual-channel high-pressure microfluidization using certain types of synthetic and natural emulsifier (Bai, Huan, Gu, & McClements, 2016; Bai & McClements, 2016a, 2016b). The current study focused on the potential of using a combination of two biosurfactants (rather than a single one) for forming and stabilizing highly concentrated emulsions. In particular, it was hypothesized that the utilization of mixed biosurfactants would improve the physical and chemical stability of the concentrated emulsions. The dual-channel homogenization method used in this study has rarely been used to form oil-in-water emulsions from natural emulsifiers, and so there is currently a relatively poor understanding of its capabilities. Nevertheless, it has some important advantages over conventional high-pressure methods for this purpose since it can continuously form emulsions in a single step, without the need to form an emulsion premix. Consequently, another important aim of this study was to elucidate the ability of dual-channel microfluidization to produce emulsions from mixed biosurfactants.

The selection of an appropriate emulsifier is one of the most important factors to consider when developing a successful emulsion-based product using any high-pressure homogenization method (Friberg et al., 2003; McClements, 2015; Stauffer, 1999). An emulsifier should rapidly adsorb to the droplet surfaces during homogenization, it should lower the interfacial tension to facilitate further droplet breakup, and it should form a protective coating to prevent droplet aggregation (Jafari et al., 2008; Santana, Perrechil, & Cunha, 2013; Walstra, 1993). Moreover, there should be enough emulsifier present to cover the surfaces of all of the droplets formed during homogenization, otherwise droplet coalescence will occur inside the homogenizer (Tcholakova, Denkov, & Danner, 2004). Many different kinds of emulsifiers are commercially available that can be used in food products, including polysaccharides, proteins, phospholipids, small molecule surfactants, and solid particles (Kralova & Sjoblom, 2009; McClements, 2015; Stauffer, 1999). However, each type of emulsifier differs in its effectiveness at producing small droplets during homogenization, and in its ability to avoid droplet aggregation under different environmental conditions, such as pH, ionic strength, heating, and freezing (Dickinson, 2003; Garti & Reichman, 1993; McClements, 2015). Food emulsifiers also vary considerably in their cost, availability, ease of use, flavor profiles, ingredient compatibility, "label friendliness", and legal status (Kralova & Sjoblom, 2009; Krog & Sparso, 2004; Stauffer, 1999). Each type of emulsifier therefore has its own advantages and disadvantages that make it suitable for particular applications.

There is particular interest in the utilization of natural plant-based emulsifiers in food products because of their perceived "label friendliness" (Dickinson, 2003; Lam & Nickerson, 2013; McClements & Gumus, 2016; Ngouemazong, Christiaens, Shpigelman, Van Loey, & Hendrickx, 2015). Quillaja saponins are biosurfactants that are highly effective at forming emulsions containing small droplets that are stable over a wide range of conditions (Ozturk, Argin, Ozilgen, & McClements, 2015; Yang, Leser, Sher, & McClements, 2013; Zhang, Bing, & Reineccius, 2015, 2016). These biosurfactants have been reported to exhibit little or no toxicity at the levels used in foods (Kensil, Soltysik, Wheeler, & Wu, 1996), but they do have a bitter taste when used at high concentrations (Ilsley, Miller, & Kamel, 2005), which may limit their application in concentrated emulsions. Moreover, a recent study showed that it was difficult to produce highly concentrated emulsions (40 or 50% oil) by dual-channel microfluidization using quillaja saponins alone (Bai et al., 2016). The objective of the current study was therefore to determine whether concentrated emulsions containing small droplets could be formed using quillaja saponins in combination with another natural surfactant (lecithin). The influence of total biosurfactant concentration and biosurfactant ratio (saponin to lecithin) on the formation and stability of concentrated (50% oil) oil-in-water emulsions produced by dual-channel microfluidization was therefore examined.

The results of this study are important for the development of highly concentrated emulsions produced from natural ingredients that can be used in food, beverage, and pharmaceutical applications.

2. Materials and methods

2.1. Materials

De-oiled soy lecithin (Ultralec®P, DL) was purchased from Archer Daniels Midland Co. (Decatur, IL), and was reported to contain 97 wt% lecithin. Quillaja saponin (Q-Naturale®, QN), which is an extract from the bark of the *Quillaja saponaria* tree, was kindly provided by Ingredion Inc. (Westchester, IL). Tween 80 (TN, MW 1310 Da, HLB 7), sodium phosphate monobasic and disodium hydrogen phosphate were provided by Sigma-Aldrich Co. (St. Louis, MO). Miglyol 812N, an example of a medium chain triglyceride (MCT), was purchased from Warner Graham Co. (Cockeysville, MD). All chemicals used were analytical grade. Double distilled and de-ionized water (Milli-Q) was used to prepare all solutions.

2.2. Emulsion preparation

Initially, an aqueous phase was prepared by weighing an amount of quillaja saponins or Tween 80 (0.25 to 2% (w/w)) into a buffer solution (5 mM phosphate buffer, pH 7.0) and stirring for 2 h to ensure dissolution. The resulting aqueous solutions were then stored overnight at 4 °C. An oil phase was prepared by dispersing 0 or 1% (w/w) of soy lecithin in MCT with heating at 50 °C for 30 min, then stirring at ambient temperature for about 2 h to ensure complete dissolution. 50 wt% oil-in-water emulsions were then prepared using a dual-channel microfluidizer (Microfluidics PureNano, Newton, MA, USA). Simultaneously, the oil and aqueous phases were fed into the homogenizer using two separate glass reservoirs, and then forced to impinge upon each other under high pressure (13,000 psi or 89.6 MPa) using a series of narrow channels.

2.3. Physical stability of emulsions

Long-term storage and accelerated tests were used to determine the physical stability of the emulsions. Aliquots (5 mL) of emulsions to which 0.02 wt% of sodium azide was added as a preservative were dispensed into a series of test tubes. The samples were then stored at 25 °C in the dark for 7 days to determine their long-term stability under ambient quiescent conditions. The stability of the emulsions to phase separation was then determined by taking digital photographs. Physical stability tests were carried out for emulsions in the absence and presence of an oil soluble dye (5 Nile red mg/mL ethanol), which was mixed with the emulsions after homogenization.

2.4. Determination of particle size and ζ potential

The particle size distribution of the emulsions was determined by static light scattering (Mastersizer 2000, Malvern Instruments Ltd., Malvern, Worcestershire, UK). Samples were diluted with buffer solution (5 mM phosphate, pH 7.0) to ensure that light waves could pass through and to prevent multiple scattering effects. The mean particle