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Effect of fish gelatin and gum arabic interactions on concentrated emulsion large amplitude oscillatory shear behavior and tribological properties

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ABSTRACT

Concentrated emulsions stabilized by proteins, polysaccharides, or a combination are the basis for many food products such as mayonnaise and heavy cream. However, fundamental influences of protein–polysaccharide interactions on concentrated emulsion tribological properties and large deformation behavior are often poorly understood, leading to difficulties in targeted development of concentrated food emulsions with palatable textures and good stability. Thus, the objective of this study was to characterize the effects of fish gelatin (FG)–gum arabic (GA) complexation on the large amplitude oscillatory shear (LAOS) and tribological behaviors of concentrated emulsions. Concentrated emulsions (oil phase volume fraction = 0.7) were prepared using FG–GA mixtures at different aqueous phase pH (3.6, 5.0, and 9.0) and characterized by rheometry and tribometry. Samples prepared with FG–GA mixtures at higher pH showed lower critical strains, increased extent of nonlinear behavior under LAOS (higher G'_3/G'_1 and G''_3/G''_1 values), increased strain hardening and shear thinning under LAOS (G'_L/G'_M and η'_L/η'_M values further from unity, respectively), and higher friction coefficients. These results were attributed to reduced FG–GA electrostatic complexation as pH increased from 3.6 to 9.0. Complexes formed at lower pH induced higher droplet monodispersity, greater network extension, and smaller oil droplets, resulting in greater structural stability to deformation. These stability differences would likely contribute to differences in food textures and stability under food processing and storage conditions.

1. Introduction

Particle–particle interactions play a key role in determining the physical properties of food emulsions. Modification and control of droplet interactions can be used to obtain food emulsions with specific stability and rheological properties (Knudsen, Øgendal, & Skibsted, 2008). Emulsion droplet stability depends largely on the emulsifier interfacial properties. Combining proteins and polysaccharides under appropriate conditions (e.g. temperature, pH, ionic strength, concentration, and protein:polysaccharide ratio) has been a successful strategy for controlling droplet interactions to produce food emulsions with specific rheological properties and improved stability (Dickinson & Hong, 1995; Dickinson, 2008a, Dickinson, 2008b; Guzey & McClements, 2006).

The shelf life of many food emulsions depends on the rheological characteristics of the component phases. For example, creaming of oil droplets in oil-in-water emulsions is strongly dependent on the viscosity of the aqueous phase. Moreover, many food emulsion sensory attributes, such as creaminess, thickness, smoothness, spreadability, and flowability, are directly related to their rheological properties (McClements, 1999). Thus, rheological measurements have been widely used as an analytical tool to provide fundamental information about the structural stabilization and interactions of biopolymers at the oil–water interface.

Rheological characteristics of emulsions have been measured by flow curves and small amplitude oscillatory shear (SAOS) in many published studies (Batista, Raymundo, Sousa, & Empis, 2006; Erçelebi & Ibanoğlu, 2009; Franco, Berjano, & Gallegos, 1997; McClements, 2004; Ng, Lai, Abas, Lim, & Tan, 2014; Silletti, Vingerhoeds, Norde, & Van Aken, 2007; Tatar, Sumnu, & Sahin, 2016). Large amplitude oscillatory shear (LAOS) testing has received increased attention in food rheological studies over the past decade because it can provide useful information on microstructural behaviors of complex food systems that cannot be

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obtained by conventional rheometry (Hyun, Kim, Ahn, & Lee, 2002). Complex materials with similar linear viscoelastic properties may exhibit different nonlinear viscoelastic behaviors (Hyun et al., 2011). Hence, LAOS measurements can provide greater insight for characterization of differences in emulsion microstructural flexibility and strength of droplet–droplet interactions. However, to the best of our knowledge, little information is available on the LAOS behavior of food emulsions.

LAOS measurements have been correlated to several food chewdown texture attributes (Melito, Daubert, & Foegeding, 2013), but rheological measurements in general are more applicable to textural attributes measured at the beginning of oral processing. Tribological (thin-film) measurements are more applicable to later stages of oral processing that involve rubbing and squeezing actions of the tongue against the palate such as creaminess, slipperiness, and smoothness (Krzeminski, Wohlhüter, Heyer, Utz, & Hinrichs, 2012). Previous studies on food tribology have focused on determining the lubrication properties of fluid foods or food components such as milk, biopolymer solutions, and emulsions stabilized with various biopolymers (Anvari & Joyner (Melito), 2017b; Bellamy, Godinot, Mischler, Martin, & Hartmann, 2009; Cambiella et al., 2006; de Hoog, Prinz, Huntjens, Dresselhuis, & van Aken, 2006; Goh, Versluis, Appelqvist, & Bialek, 2010). Liu, Stieger, van der Linden, and van de Velde (2015) found that emulsion lubricity can be modified by altering emulsion stability, e.g. changing protein content, fat type, and droplet–matrix interactions. Protein–polysaccharide interactions at the oil–water interface play an important role on emulsion formation and stability. Nevertheless, limited information is available on the effect of biopolymer interactions on food emulsion tribological properties.

In a previous study, we considered the influence of fish gelatin–gum arabic complexation at different aqueous phase pH (3.6, 5.0, and 9.0) on the formation and stability of concentrated emulsions (Anvari & Joyner (Melito), 2017a). Electrostatic associative complexation between fish gelatin and gum arabic resulted in insoluble complexes at pH = 3.6, yielding concentrated emulsions with higher stability to creaming and larger viscoelastic moduli values under SAOS. However, because most food processing and oral processing operations involve large strains and sliding actions, additional study on nonlinear viscoelastic properties and tribological behavior is needed for a more comprehensive evaluation of the influenced of biopolymers interactions on concentrated emulsions structure–function relationships. Accordingly, the objective of this study was to characterize the effects of fish gelatin–gum arabic complexation at different aqueous phase pH on concentrated emulsion tribological and LAOS behaviors.

2. Materials and methods

2.1. Materials

Fish gelatin from cold water fish skin (Type B, pI = 4.81 (Yang, Anvari, Pan, & Chung, 2012)), gum arabic, and sodium azide (NaN_3) were purchased from Sigma Chemical Co. (St. Louis, MO, USA). Corn oil was purchased from a local supermarket (Moscow, ID, USA).

2.2. Preparation of fish gelatin–gum arabic aqueous mixtures

The procedure developed by Yang et al. (2012) was used to prepare the fish gelatin–gum arabic aqueous mixtures. Briefly, fish gelatin and gum arabic were dissolved separately in distilled water at 40 °C, then mixed at a 1:1 weight ratio and 2% (w/v) total biopolymer concentration. The pH was adjusted to 3.6, 5.0, or 9.0 using 1 M sodium hydroxide or 1 M acetic acid; sodium azide (0.2% w/v) was added as a preservative. Prepared mixtures were incubated for 24 h in a shaking water bath (100 rpm) at 25 °C to promote electrostatic attractive interactions.

2.3. Emulsion preparation

To prepare the concentrated emulsions, the fish gelatin–gum arabic aqueous mixtures at different pH were added to the oil phase (final oil phase volume fraction of 0.7) and homogenized using a Polytron stand homogenizer (Kinematica AG, NY, USA) at 10,000 rpm for 60 s at room temperature (25 ± 2 °C).

2.4. Rheological measurements

Rheological measurements were conducted with a Discovery Hybrid Rheometer (DHR-3, TA Instruments Inc., New Castle, DE, USA) at 25 °C using a cone and plate geometry (1° angle, 40 mm diameter) equipped with a solvent trap. Strain sweeps (0.01–100%, 6.28 rad/s) were used to determine concentrated emulsion critical strains and properties at critical strain. LAOS testing was conducted by collecting raw waveform data at 0.5, 50, and 100% strains at three different frequencies (0.5, 1, and 10 rad/s). All measurements were performed in triplicate.

2.5. Tribological measurements

Tribological measurements were conducted on a Discovery Hybrid Rheometer (DHR-3, TA Instruments Inc., New Castle, DE, USA) at 25 °C using a double-ball (polypropylene balls; McMaster-Carr, Atlanta, USA) geometry on 40 mm diameter polydimethylsiloxane (PDMS) gel plates. PDMS plates were prepared using a two-part silicone elastomer kit (Sylgard 182, Dow Corning Corporation, Midland, USA) with a 10:1 w/w ratio of elastomer base to curing agent according to the method of Anvari and Joyner (Melito) (2017b).

For each tribological test, approximately 3 mL of sample was placed on the plate and the upper tool was lowered to 1 N normal force. The normal force was selected to mimic the in-mouth force during oral processing, which is 0.01–10 N (Miller & Watkin, 1996). After reaching 1 N normal force, sliding speed was increased stepwise from 0.1 to 1000 mm/s and the resulting friction coefficients recorded. Testing surfaces were cleaned with 70% ethanol and dried with laboratory wipes before each test. All tribological testing was conducted in triplicate.

Torque and spindle rotational speed were converted to friction coefficient and sliding speed, respectively, using Equations (1) and (2):

$$v = R\omega \tag{1}$$

$$\mu = M/(RF_N) \tag{2}$$

where ν is sliding speed (m/s), R is the radius from the pivot point to the ball (m), ω is spindle rotational speed (rad/s), F_N is normal force (N), μ is friction coefficient (unitless), and M is torque (N m).

2.6. Data analysis

Strain sweep data were analyzed using ANOVA followed by Tukey's test using SAS 9.3 (SAS Institute, Cary, NC, USA). LAOS data were analyzed using the MITlaos program (MITlaos beta) developed by Ewoldt, Clasen, Hosoi, and Mckinley (2007) designed for MATlab (MathWorks Natick, Massachusetts, USA).

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