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Gum Arabic-stabilized conjugated linoleic acid emulsions: Emulsion properties in relation to interfacial adsorption behaviors

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ABSTRACT

Conjugated linoleic acid (CLA) is a dietary supplement due to its multiple health benefits. However, significant loss of biologically active CLA could occur due to oxidation. Oil-in-water emulsion is supposed to be an effective protection system. In a previous study (Yao et al., 2013), physical and chemical stabilities of CLA emulsion stabilized with gum Arabic were investigated, and the optimal gum concentration was identified as 5 wt%. The present study dealt with the interfacial adsorption of three gum Arabic samples (one conventional gum, GA; two matured gums, EM2 and EM10) at the CLA-water interface, in relation to their emulsifying properties. With increasing gum Arabic concentration, particle size ($D[3,2]$) of CLA emulsion decreased and slowly leveled off, which was accompanied with a gradual saturation of gum Arabic adsorbed at the emulsion interface. Nevertheless, surface load of gum Arabic at the emulsion interface (Γ, mg/m^2) reached maximum at a gum concentration of 5 wt%. This could reasonably explain the optimal emulsion stability observed at this gum concentration, as revealed by particle size distribution of fresh emulsions and by acceleration test at 60 °C. Among the three gums, EM10 exhibited the highest emulsifying activity and conferred the best emulsion stability, despite of its lowest surface load. This is presumably due to a larger tendency of the AGP fraction in EM10 toward aggregation at the CLA-water interface.

1. Introduction

Conjugated linoleic acid (CLA), a mixed group of positional and geometrical isomers of linoleic acid (C18: 2) (Khaskheli, Talpur, Demir, Cebeci, & Jawaid, 2013), has been a subject of growing interest due to its nutritional and therapeutic properties (Villeneuve et al., 2007). A series of scientific studies has shown that CLA has many potential health benefits, such as cancer prevention, anticarcinogenic activity, weight control, and bone formation (Lock & Bauman, 2004). However, CLA is not stable during thermal processing and significant loss of biologically active CLA occurs through oxidation. It is important to develop an effective protection system for delivery of CLA in food products (Campbell, Drake, & Larick, 2003; Herzallah, Al-Ismail, & Humeid, 2005).

Lipids are often present in food products in the form of oil-in-water (O/W) emulsions (Charoen et al., 2012). In this study we examine how O/W emulsions stabilized with food hydrocolloids can be used as an effective delivery system to encapsulate and protect CLA. Emulsions are thermodynamically unstable system from a physicochemical point of view, rapidly or slowly separating into two immiscible phases over a period of time (Borwankar, Lobo, & Wasan, 1992) and are stabilized by improvement of their kinetic stability (Bergenstahl & Claesson, 1997), where stability may be defined as the resistance to physical changes. Meanwhile, the stability of emulsion can be related to a kinetic concept, because the long-term stability of a dispersed system implies that the rate and extent of change in its structure and properties are sufficiently low in real time (Tolstoguzov, 1995). O/W emulsions consist of small oil droplets dispersed in an aqueous medium, with each droplets being coated by a thin layer of emulsifier molecules (McClements, 2012),

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including proteins, polymers, ionic and non-ionic surfactants (Dickinson, 2001). They stabilize the emulsion by their ability to generate repulsive interactions (steric and electrostatic) between oil droplets (McClements, 2004). Most hydrocolloids can act as stabilizers (stabilizing agents) of O/W emulsions, but only a few can act as emulsifiers (emulsifying agents). The latter functionality requires substantial surface activity at the O/W interface, and hence the ability to facilitate the formation and stabilization of fine droplets during and after emulsification (Dickinson, 2003). Emulsifiers that adsorb at the O/W interface influence noticeably lipid oxidation (Genot, Meynier, Riaublanc, & Kamal-Eldin, 2003; Waraho, McClements, & Decker, 2011).

Gum Arabic, a mixture of anionic polysaccharides and protein fraction (Al-Assaf & Phillips, 2008), derived from the natural bark exudates of Acacia trees, is the most commonly used biopolymer emulsifier. Gum Arabic is an effective emulsifier by virtue of its high water solubility, low solution viscosity, good surface activity, and ability to form a protective film around emulsion droplets. It is recognized by many researchers that gum Arabic consists of three major components: arabinogalactan (AG, ~90% of total mass), arabinogalactan protein (AGP, ~10% of total mass), and glycoprotein (GP, ~1% of total mass) (Randall, Phillips, & Williams, 1989; Renard, Lavenant-Gourgeon, Ralet, & Sanchez, 2006). The AGP represents ~10% protein, and a 'wattle blossom-type' structure has been suggested, in which carbohydrate blocks are linked to a common polypeptide chain (Mahendran, Williams, Phillips, Al-Assaf, & Baldwin, 2008). The existence of AGP provides an excellent interfacial property for gum Arabic, which is attributed to the 'wattle blossom-type' structure providing both hydrophobic polypeptide chain and hydrophilic carbohydrate blocks, conferring good emulsification characteristics (Castellani et al., 2010). The AGP fraction is bound to carbohydrate, adsorbing onto the O/W interface as an anchor, and the carbohydrate portion contributes mainly to emulsion stability by steric repulsion, preventing flocculation of the oil droplets by forming a hydrated layer (Nakauma et al., 2008).

A maturation process is employed to modify gum Arabic by heating at an elevated temperature and controlled humidity, resulting in the increase of AGP fraction and thus enhanced emulsifying ability (Yao, et al., 2013). EM2 and EM10 are two commercial gum Arabic samples modified by the maturation process. The time required for heat treatment to obtain EM10 is longer than that for the production of EM2 (Castellani et al., 2010), thus a high AGP content for EM10.

The previous work in our laboratory (Yao et al., 2013) has demonstrated that the three gums could produce physically and chemically stable CLA O/W emulsion. The present study is to investigate in more details the emulsion properties in relation to the interfacial adsorption behaviors of gum Arabic at CLA-water interface.

2. Materials and methods

2.1. Materials

Conventional commercial GA and two matured gums (EM2 and EM10) in spray-dried form were provided by San-Ei Gen F.F.I., Japan. The three gum samples were used without further purification. Food grade CLA with a purity of 80% was purchased from Beijing Health Science and Technology Co. Ltd., China. The remaining components contain 12.5% of oleic acid, and palmitic acid, stearic acid, and linoleic acid account for the rest, which were measured by a gas chromatograph (Varian 3900, USA). The density of CLA was 0.9050 g/mL. Distilled and deionized water were used for the preparation of all solutions.

2.2. Solution preparation

Gum Arabic solutions were prepared at the corresponding concentration by dispersing a weighed amount of gum Arabic powder in distilled water, and stirred at 25 ± 0.2 °C overnight to ensure the complete dissolution.

2.3. Emulsion preparation

Emulsion was prepared by blending 15.0 wt% oil phase (CLA) with 85 wt% aqueous phase (containing 2.5, 5.0, 7.5, 10.0, or 15.0 wt % gums) using a high-speed PT-MR2100 Polytron-type mixer (Kinematica Co., Switzerland) at 26,000 rpm for 3 min, followed by one pass through the high-pressure homogenizer (Microfluidics M-110L, USA) at 75 MPa. The whole process was carried out in an ice bath to minimize any possible lipid oxidation.

2.4. Emulsion characterization

2.4.1. Particle size analysis

The physical stability of emulsion was evaluated by using a storage acceleration test at 60 °C for a period of 7 days (Aoki, et al., 2007; Li, Fang, Al-Assaf, Phillips, & Jiang, 2012). The particle size and particle size distribution (PSD) of the emulsions were measured using a laser light scattering instrument (MasterSizer 2000, Malvern instruments Ltd.). The emulsion sample was dispersed in recirculating water in measuring cell until an obscuration rate of 10–20% was obtained. The refractive index values used for dispersed and continuous phases were 1.52 and 1.33, respectively. The absorption coefficient was 0.01. The average droplet diameters of the emulsions were determined as $D[3,2]$ and $D[4,3]$, representing the surface-volume mean diameter and the volume-moment mean diameter, respectively. Each test was repeated in triplicate at 25 ± 0.2 °C.

2.4.2. ζ-potential measurements

The eletrophoretic mobility expressed as ζ-potential was determined using a Zetasizer Nano-ZS (Malvern Instruments, UK). The emulsions were diluted approximately 1000 times using deionized water to avoid multiple scattering effects before measurements at 25 ± 0.2 °C. Data on the ζ-potential was presented as mean ± SD of triplicate.

2.4.3. Steady shear viscosity

The rheological measurements were performed using a Haake Rheostress6000 (Thermo Fisher Scientific, Germany). A cone-and-plate geometry (cone diameter 60 mm, angle 1°, gap 0.052 mm) was employed in all measurements. The steady shear viscosity was measured at shear rates of 0.01–500 s^{-1}. The temperature was maintained at 25 ± 0.2 °C.

2.4.4. Adsorption quantity on droplet interface

The interfacial adsorption quantity on emulsion droplet surface was determined according to the method of Nakauma et al. (2008). Fresh emulsion (5 g) was mixed with a solution A (20 g, 36 wt% sugar, 6 wt% NaCl and 58% water) using an Ika-Eurostar mixer (Staufen, Germany) at 200 rpm for 1 min. The mixture was centrifuged at 10,000 g for 30 min at 10 °C to recover the lower aqueous phase. The residue (i.e., intermediate creaming phase and the upper oil phase) was treated three times with the solution A (total weight 25 g) using the same protocol to recover the lower aqueous phase. The final residue was mixed with a solution B (25 g, 0.1 wt% SDS, 5 wt% NaCl, and 94.9% water) using a PT-MR2100 Polytron-type mixer (Kinematica Co., Switzerland) at 10,000 rpm for 2 min. The role of surfactant SDS here is to displace the materials