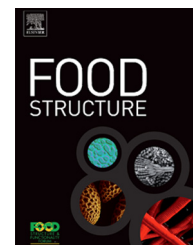


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Review

Structure–function relationships in food emulsions: Improving food quality and sensory perception

Cheryl Chung, David Julian McClements*

Department of Food Science, University of Massachusetts, Amherst, MA 01003, United States

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ABSTRACT

Increasing consumer demand for higher quality, cheaper, more convenient, and healthier emulsion-based products means that the food industry must have a good understanding of the relationship between the structural and functional properties of food emulsions. This review article provides an overview of the relationship between the composition and structural organization of oil-in-water emulsions and their physicochemical (optical, rheological, and stability) and sensory (appearance, texture, flavor, and mouthfeel) properties. It also discusses recent advances in the design of structured emulsions with novel functional properties, such as multiple emulsions, filled-hydrogel particles, multilayer emulsions, microclusters, and air-filled emulsions.

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* Corresponding author. Tel.: +1 413 545 1019; fax: +1 413 545 1262.

E-mail address: mcclements@foodsci.umass.edu (D.J. McClements).

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1. Introduction

Many food products contain two immiscible phases (typically oil and water) as part of the ingredients, and it is crucial to mix and stabilize them well in order to produce high quality, stable, and sensory appealing products. These two immiscible phases are often incorporated into food products as emulsions, which are formed by dispersing one phase into the other in the form of small droplets (McClements, 2005; Santana, Perrechil, & Cunha, 2013; Windhab, Dressler, Feigl, Fischer, & Megias-Alguacil, 2005). A wide variety of food ingredients and products can be considered to consist either entirely or partially as emulsions, or have been in an emulsified state sometime during their production, *e.g.*, beverages, butter, cheese, colorants, cream, desserts, flavors, ice cream, margarines, milk, salad dressings, sauces, soups, and yogurts (Leal-Calderon, Thivilliers, & Schmitt, 2007; McClements, 2010b; Mendez-Velasco & Goff, 2011; Moore, Duncan, Rasor, & Eigel, 2012; Sanguansri et al., 2013). The emulsified components of these foods play important roles in determining their distinct functional attributes, such as appearance, texture, stability, and flavor (Benjamins, vingerhoeds, Zoet, de Hoog, & van Aken, 2009; Chojnicka-Paszun, de Jongh, & de Kruif, 2012; Chung, Olson, Degner, & McClements, 2013d; Aken, Vingerhoeds, & de Wijk, 2011; Vliet, van Aken, de Jongh, & Hamer, 2009).

Conventionally, emulsions are classified as either oil-in-water (O/W) emulsions in which oil forms the dispersed phase (oil droplets) and water forms the continuous phase (*e.g.*, milk and sauces), or water-in-oil (W/O) emulsions in which water forms the dispersed phase and oil forms the continuous phase (*e.g.*, butter and margarines) (Fig. 1) (McClements, 2005). Emulsions can also be classified according to their droplet sizes: *nanoemulsions* with droplet radii ranging from around 10 to 100 nm; *miniemulsions* with droplet radii ranging from about 100 to 1000 nm; and *macroemulsions* with droplet radii from about 1000 nm to 1000 μm (McClements, 2012b; Santana et al., 2013; Windhab et al., 2005). However, in reality there is no distinct change in physicochemical properties at the boundaries between these different systems. Structurally complex emulsions with enhanced functional properties can often be formed using advanced processing methods, such as oil-in-water-in-water ($\text{O}/\text{W}_1/\text{W}_2$) emulsions, water-in-oil-in-water ($\text{W}_1/\text{O}/\text{W}_2$) emulsions, and oil-in-water-in-oil ($\text{O}_1/\text{W}/\text{O}_2$) emulsions (Fig. 1) (Dickinson, 2011; McClements, 2012a; Muschiolik, 2007). In this article, we mainly focus on the formation and properties of oil-in-water emulsions since these are the most widely used in the food industry, but we also highlight some new developments using structurally complex emulsions due to their potential for certain food applications.

The various components within a food emulsion may significantly affect its bulk physicochemical properties and

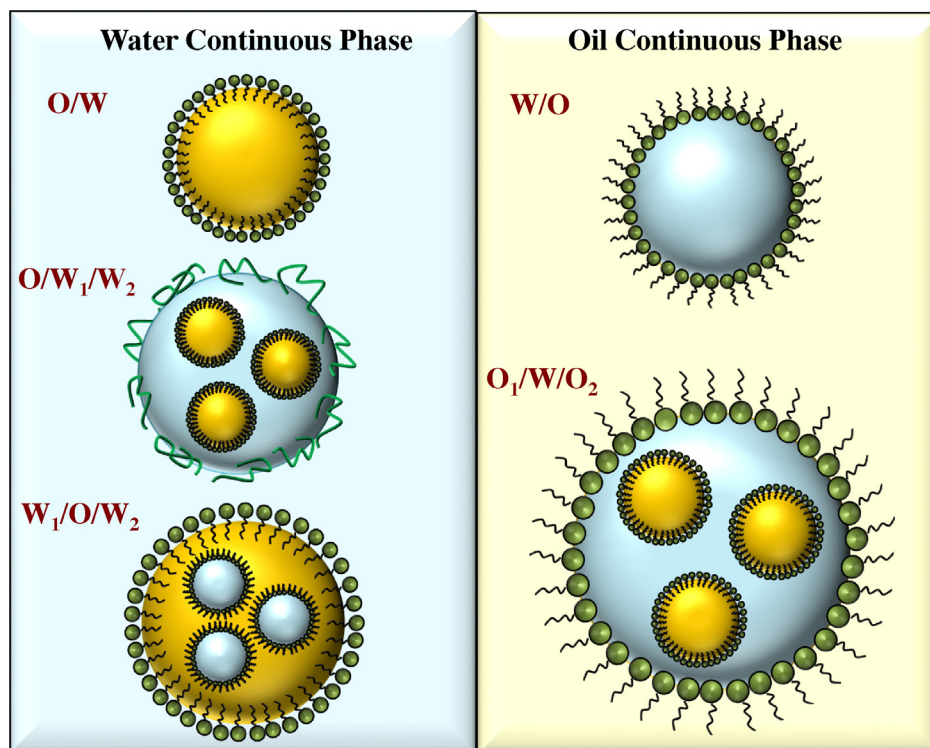


Fig. 1 – Schematic diagram of simple emulsions – oil-in-water (O/W) emulsion, and water-in-oil (W/O) emulsion, and multiple emulsions – oil-in-water-in-water (O/W/W) emulsion, water-in-oil-in-water (W/O/W) emulsion, and oil-in-water-in-oil (O/W/O) emulsion.

sensory attributes (Fig. 2). In this review, we discuss the various structural elements present in oil-in-water (O/W) emulsions and their role in determining the overall properties of emulsion-based food products. Initially, the influence of emulsion components on the physicochemical and sensory properties of emulsions is discussed. An overview of some of the instrumental methods commonly used to establish the key factors that influence the properties of food emulsions is then given. In the final section, we briefly review some recently developed structural design approaches that can be used to create emulsions with novel functional attributes e.g., low fat products with similar sensory qualities as the full fat counterparts.

2. Emulsion system formation

Kinetically stable emulsions are formed using amphiphilic compounds (“emulsifiers”) that facilitate emulsion formation and improve emulsion stability (Fig. 1) (Dickinson, 2009b; McClements, 2005; Santana et al., 2013). After droplet formation, emulsifiers form a thin coating around the oil droplets that inhibits their aggregation by generating repulsive forces between them. Emulsifier characteristics (type and concentration), bulk phase properties (interfacial tension and viscosity) and shearing conditions (pressure, number of passes, and instrument type) affect the characteristics of the oil droplets produced (size, charge, interactions, and

organization), as well as the bulk physicochemical properties of the emulsions (rheology, appearance, flavor, and physical and chemical stability) (Charoen et al., 2012; Chung, Degner, & McClements, 2012b; Huck-Iriart, Candal, & Herrera, 2011; Perrechil & Cunha, 2010; Seta, Baldino, Gabriele, Lupi, & de Cindio, 2012; Zinoviadou, Moschakis, Kiosseoglou, & Biliaderis, 2011).

Emulsifiers commonly used in food applications include low molecular weight surfactants, proteins, phospholipids, polysaccharides and their derivatives (Dickinson, 2006, 2009b; Horn, Nielsen, & Jacobsen, 2012; Kravola & Sjoblom, 2009; Seta et al., 2012; Singh, 2011). The choice of a specific emulsifier (or blend of emulsifiers) for a particular application is critical for creating successful products, and depends on product formulation, processing conditions, and the desired final properties (Horn et al., 2012; McClements, 2005; Seta et al., 2012). In addition to oil, water and emulsifier, most food emulsions also contain other ingredients to improve their texture, stability, and taste, such as thickeners, gelling agents, weighting agents, ripening inhibitors, flavors, and colorants (Dickinson, 2006; Dolz, Hernandez, Delegido, Alfaro, & Munoz, 2007; Perrechil & Cunha, 2010; Su, Lien, Lee, & Ho, 2010).

The fundamental steps in emulsion formation have been reviewed in detail elsewhere (McClements, 2005; Santana et al., 2013). After formation, an emulsion contains a number of different regions: the *dispersed phase*, the *continuous phase*, and the *interfacial layer* (Fig. 1). The properties and structural organization of these various components within a food

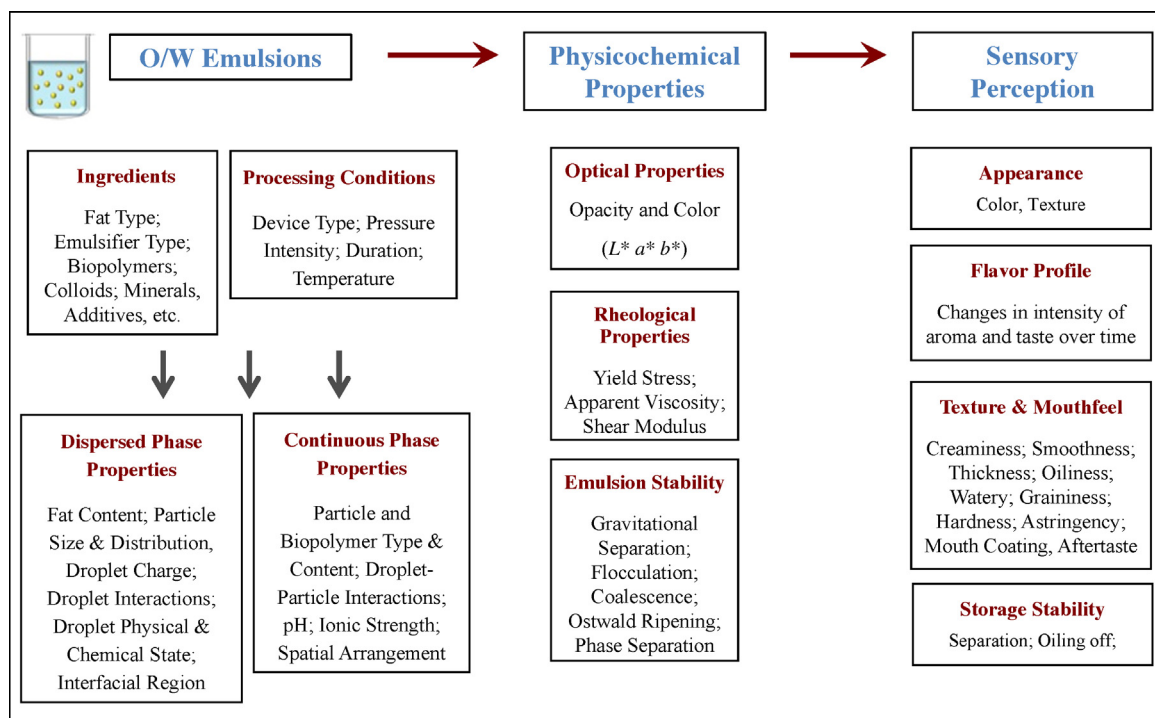


Fig. 2 – Overview of the relationship between oil-in-water emulsion properties and the bulk physicochemical properties and sensory characteristics of food emulsions.

product plays an important role in determining its physicochemical and sensory properties, which will be discussed in the following sections.

3. Food emulsion building blocks

3.1. Dispersed phase

In this section, we will briefly highlight the major characteristics of the dispersed phase that influences the properties of O/W emulsion systems.

Droplet concentration – The amount of oil present within an emulsion has a major effect on its physicochemical and sensory properties *e.g.*, optical, rheological and stability properties. The oil droplet concentration can vary from a fraction of a percent in beverage emulsions (McClements, 2005) to over 70% in mayonnaises (de Wijk & Prinz, 2007; Depree & Savage, 2001).

Particle size distribution – The size of the droplets in an emulsion also plays a critical role in determining its physicochemical properties. The average droplet size and particle size distribution (PSD) of emulsions can be controlled by varying processing conditions (*e.g.*, homogenizer type, pressure and/or duration, and temperature) (McClements, 2005; Santana et al., 2013) and/or system formulation (*e.g.*, oil, emulsifier, salts, sugars, cosolvents and other solutes) (Cerimedo, Iriart, Candal, & Herrera, 2010; Huck-Iriart et al., 2011; McClements, 2005). The PSD of an emulsion can be measured by various particle-sizing technologies (such as dynamic light scattering, static light scattering, or electrical pulse counting)

or it can be determined by various microscopy methods (such as optical or electron microscopy).

Droplet charge – The droplets in most food emulsions have an electrical charge that depends on the type of emulsifiers used (McClements, 2005) and solution conditions (such as pH, ionic strength and polyelectrolytes) (Harnsilawat, Pongsawatmanit, & McClements, 2006; McClements, 2005; Tippetts & Martini, 2012). The electrical characteristics of oil droplets are usually characterized in terms of the ζ -potential versus pH profile (Hunter, 1986). Droplet charge has a major influence on droplet interactions. For example, two droplets with similar charge signs (*i.e.*, both negative or both positive) will repel each other whereas two oppositely charged droplets will attract each other (Mao & McClements, 2011, 2012). Overall, the electrical charges on oil droplets and the resulting interactions with the surrounding droplets and other ingredients significantly influence the physical and chemical properties of food emulsion systems (see later).

Droplet interactions – Droplets in an emulsion may interact with each other and with other components through a variety of molecular and colloidal interactions, such as van der Waals, electrostatic, steric, depletion, hydrogen bonding, and/or hydrophobic interactions (McClements, 2005). These interactions are influenced by droplet characteristics (*e.g.*, size, dielectric constant, and refractive index), interfacial properties (*e.g.*, charge, thickness, polarity, and packing), and continuous phase environment (*e.g.*, dielectric constant, refractive index, pH, and ionic strength). Depending on the relative strength of the attractive and repulsive interactions, oil droplets can merge together (coalesce), associate with each other (flocculate), or remain as individual entities. Droplet-droplet interactions have

a major impact on the physicochemical properties and stability of emulsions. Droplets may also interact with various other components present within emulsions, *e.g.*, minerals, proteins, or polysaccharides (Guzey & McClements, 2007; Tran & Rousseau, 2013).

3.2. Continuous phase

The continuous phase of emulsion-based food products may contain a variety of components, such as salts, sugars, acids, bases, buffers, starch granules, hydrocolloids, particulates, air bubbles, and flavor compounds. Many of these components influence the structure (Fig. 3), physicochemical properties, and sensory attributes of emulsions. For example, hydrocolloids, starch granules and air bubbles greatly increase the viscosity and influence the other textural attributes of emulsions (Cerimedo et al., 2010; Chung et al., 2012b; Huck-Iriart et al., 2011; Perrechil & Cunha, 2010; Protonotariou, Evageliou, Yanniotis, & Mandala, 2013). Therefore, it is important to establish the physicochemical characteristics of the various components within an emulsion-based product to better understand how they contribute to the overall properties of the system. Some of the most important characteristics that should be established are composition, concentration, size, shape, charge, polarity, interactions, density, refractive index, and physical state.

3.3. Interfacial region

The interfacial region consists of the thin boundary layer that separates the oil phase from the water phase (Fig. 1). This layer typically consists of a mixture of emulsifier, oil, and water molecules, with other components also possibly being present, such as mineral ions, hydrocolloids, antioxidants, and prooxidants. Although the interfacial region only occupies

a small volume of the overall system, its properties can have a pronounced influence on the physicochemical and sensory properties of food products, including their rheology, stability and flavor (Christiansen et al., 2011; Dickinson, 2001). Hence, manipulation of interfacial characteristics is usually one of the most effective methods in designing food products with specific functional performances (Dickinson, 2003).

4. Sensory properties of food emulsion products

The composition and structural organization of emulsions ultimately determine their desirable sensory attributes. In this section, we therefore focus on how the physicochemical and sensory properties of emulsions are influenced by *dispersed phase*, *continuous phase*, and *interfacial region* characteristics (Fig. 2) (Akhtar, Stenzel, Murray, & Dickinson, 2005; Benjamins et al., 2009; Chojnicka-Paszun et al., 2012; Chung, Olson, et al., 2013; van Aken et al., 2011; van Vliet et al., 2009). The initial sensory perception of a food product, such as appearance, texture, and aroma, depend on the initial properties of a food emulsion. However, the behavior of a food emulsion within the mouth during mastication also plays a major role in its sensory perception due to changes in its composition and structure brought about by chewing, dilution with saliva, and surface coating (Cakir et al., 2012; Ranc et al., 2006; Togashi, Morita, & Nakazawa, 2000).

In general, the term “flavor” has been used to refer to the combined perception of aroma, taste, mouthfeel, and texture (British Standards Institute, 1975). However, consumers mainly associate this term with aroma and taste. In this review, we use the term “flavor profile” to refer to aroma and taste attributes, and “texture” and “mouthfeel” to refer to those sensations that arise from the tactile stimuli in the tongue and palate during mastication.

4.1. Optical properties and appearance

The main optical properties of emulsions are their *opacity* and *color*. The overall appearance of emulsions depends on their composition and microstructure (Chung et al., 2012b; Chung, Degner, & McClements, 2013c; McClements, 2002, 2005). The opacity and color of emulsions are mainly determined by particle concentration, size, and refractive index contrast, as well as the presence of any chromophores that absorb light.

4.1.1. Dispersed phase characteristics

The perceived lightness of an emulsion increases with increasing oil content, especially from 0 to 5% fat, due to increased light scattering by the fat droplets (Fig. 4) (Chantrapornchai, Clydesdale, & McClements, 1999; Chung et al., 2012b; McClements, 2002). This phenomenon may have important implications for the creation of reduced-fat food products, since decreased lightness may be associated with undesirable loss of “creaminess” (McClements, 2002). The decrease in lightness of reduced fat products due to reduction of fat droplet concentration can often be compensated for by optimizing formulation and/or processing conditions. For example, the particle size distribution (PSD) can be controlled

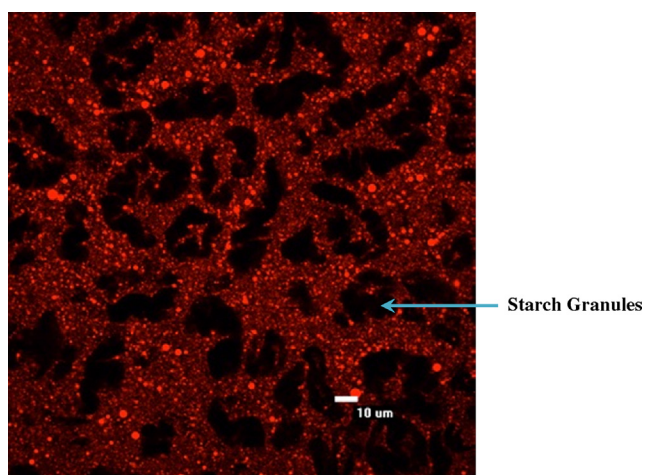


Fig. 3 – Confocal micrograph (60×) of mixed system containing 8% oil-in-water emulsion and 3.5% swollen starch granules (~38 μm). The dark spots are starch granules (unstained) while the red spots are stained oil droplets. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

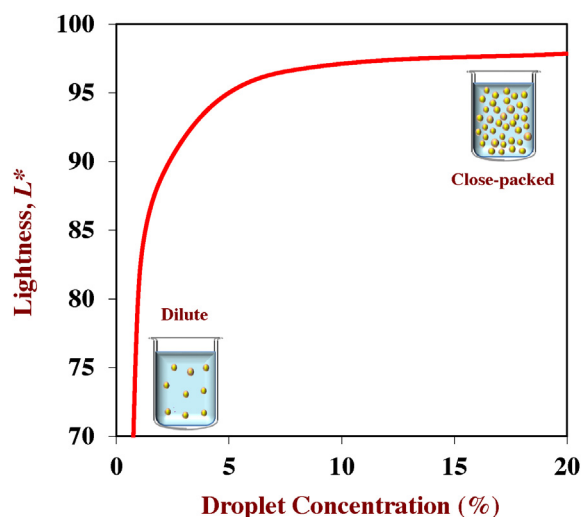


Fig. 4 – Dependence of the lightness of oil-in-water emulsions on droplet concentration.

so as to increase the light scattering efficiency of the oil droplets by fabricating droplets with sizes similar to the wavelength of light (~ 500 nm). Droplet sizes below and/or above this optimal size scatter lesser light and hence give lower lightness (McClements, 2002).

4.1.2. Continuous phase characteristics

Various components in the continuous phase can also alter the optical properties of emulsions due to their ability to scatter or absorb light waves. For particulate matter (such as starch granules, protein particles, or air bubbles) the magnitude of this effect depends on their relative refractive indices, concentrations, and sizes (McClements, 2002, 2005). Incorporation of non-fat particles that scatter light in a similar manner to oil droplets increases the opacity of emulsions, and hence may be a potential alternative for reduced-fat products, e.g., titanium dioxide (Chantrapornchai, Clydesdale, & McClements, 2000) and protein microparticles (Ako, Nicolai, & Durand, 2010; Dissanayake, Kelly, & Vasiljevic, 2010; Gulzar, Lechevalier, Bouhallab, & Croguennec, 2012; Kennedy, Mounsey, Murphy,

Duggan, & Kelly, 2006). The presence of water-soluble colorants in the continuous phase will also influence the appearance of an emulsion due to their ability to selectively absorb light waves. The color of emulsions will also be influenced by any chromophores dissolved within the oil phase.

Addition of components that alter the structural organization, and therefore light scattering pattern, of fat droplets may also influence the appearance of emulsion-based products. For example, addition of locust bean gum (LBG) to emulsions promoted droplet aggregation, which reduced their lightness (Fig. 5) (Chung, Degner, & McClements, 2013a). Alternatively, novel structural design principles can be used to create reduced fat products with high opacity, e.g., hydrogel particles (Chung, Degner, & McClements, 2013b; Zhang et al., 2013). More details on these structural design approaches and their potential use in food product development are given in Section 6.

4.1.3. Interfacial region

The interfacial region would not be expected to have a large direct effect on the optical properties of an emulsion because its thickness (δ) is much smaller than the wavelength of light ($\delta \ll \lambda$). In addition, in most conventional emulsions the volume occupied by the interfacial region only makes up a small contribution to the overall particle volume (McClements, 2011), and hence to the overall optical properties. Nevertheless, the optical properties of the interfacial region may be important in nanoemulsions, since then it makes up an appreciable contribution to the overall particle volume. In this case, information about the thickness and composition of the interfacial region are needed, as well as knowledge of its refractive index and absorption properties. The properties of the interfacial region may also have an indirect effect on the optical properties of emulsions-based products by altering the aggregation state of the droplets, and therefore their light scattering efficiency.

4.2. Flavor profile

The overall flavor profile of a food emulsion depends on the distribution of flavor molecules amongst the various phases present (e.g., oil, water, interface, headspace), and their release

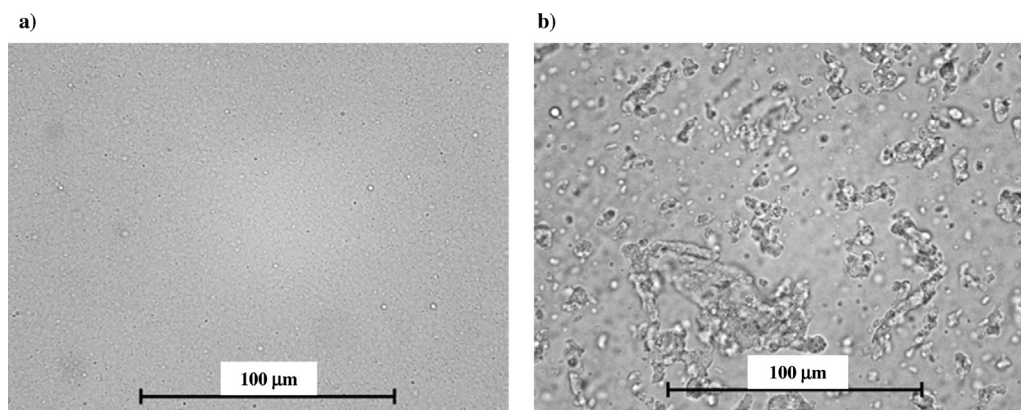


Fig. 5 – Optical micrographs (60 \times) of (a) oil-in-water emulsion (5% oil) and (b) mixed systems of locust bean gum (0.4%) and oil-in-water emulsion (5% oil).

profile during consumption. The release of flavor molecules from food emulsions is determined by their equilibrium partition coefficients and their mass transport kinetics (Frank, Appelqvist, Piyasiri, Wooster, & Delahunty, 2011; Karaishou, Blekas, & Paraskevopoulou, 2008; Leksrisonpong, Barbano, Foegeding, Gerard, & Drake, 2010; McClements, 2005). Flavor release is usually characterized by an increase in the concentration of flavor molecules in the aqueous phase (taste) or headspace (aroma) as a function of time (flavor intensity–time relationship) (McClements, 2005; Seuvre, Philippe, Rochard, & Voilley, 2007).

4.2.1. Dispersed phase characteristics

The flavor profile of emulsion-based foods depends on various oil droplet properties, e.g., oil type, concentration, size, and physical state. Fats and oils from different sources (e.g., corn oil, olive oil, vegetable oil, and animal fats) differ in the type and concentration of volatile compounds they contain, and hence contribute differently to the flavor profile of foods (Cunha, Grimaldi, Alcantara, & Viotto, 2013; Vingerhoeds, de Wijk, Zoet, Nixdorf, & van Aken, 2008; Vitova, Loupanova, Sklenarova, Divisova, & Bunka, 2012).

The fat droplet concentration influences the perception of volatile and non-volatile flavor compounds by altering their partitioning between the oil, water, and headspace phases (Fig. 6a). With increasing fat content, the concentration of non-polar flavors in the headspace above an emulsion is reduced, and therefore their perceived flavor intensity decreases (Bayarri, Smith, Hollowood, & Hort, 2007; Bayarri, Taylor, & Joanne, 2006; Christiansen et al., 2011; Leksrisonpong et al., 2010; Mao, Roos, & Miao, 2013; Weel et al., 2004), whereas the opposite is true for polar flavors (Fig. 6a). This phenomenon is particularly important to take into account when designing reduced-fat products.

The flavor intensity of full fat products may be characterized as balanced and sustainable throughout the course of consumption (“sustained release”), whereas in reduced fat products the flavor intensity may be unbalanced due to an initial spike (“burst release”) of flavor immediately after consumption, followed by low flavor intensity at later times (Fig. 6b) (Frank, Eyres, Piyasiri, & Delahunty, 2012; Ma & Boye, 2013; Malone & Appelqvist, 2003). The initial flavor burst in reduced fat products is due to an appreciable fraction of the non-polar flavor molecules being present in the water phase rather than in the oil phase (Malone & Appelqvist, 2003). The composition and structure of reduced-fat products may therefore have to be redesigned so that their release profiles more closely match those of high fat products. Sustained release may be achieved by encapsulating non-polar flavor molecules in delivery systems, such as filled hydrogel particles or microencapsulated particles. As shown in Fig. 7, the individual oil droplets (1%, w/w) in conventional oil-in-water emulsion are distributed randomly while those in the hydrogel particle systems are embedded in the hydrogel particle matrix. With the entrapment of the oil droplets the release of flavor compounds to the headspace can be delayed and sustained due to the longer diffusion path-length of the hydrogel particles.

Fat droplet size may also influence the kinetics of flavor release for non-polar flavor compounds since they must diffuse out of the oil droplets before being released into the aqueous

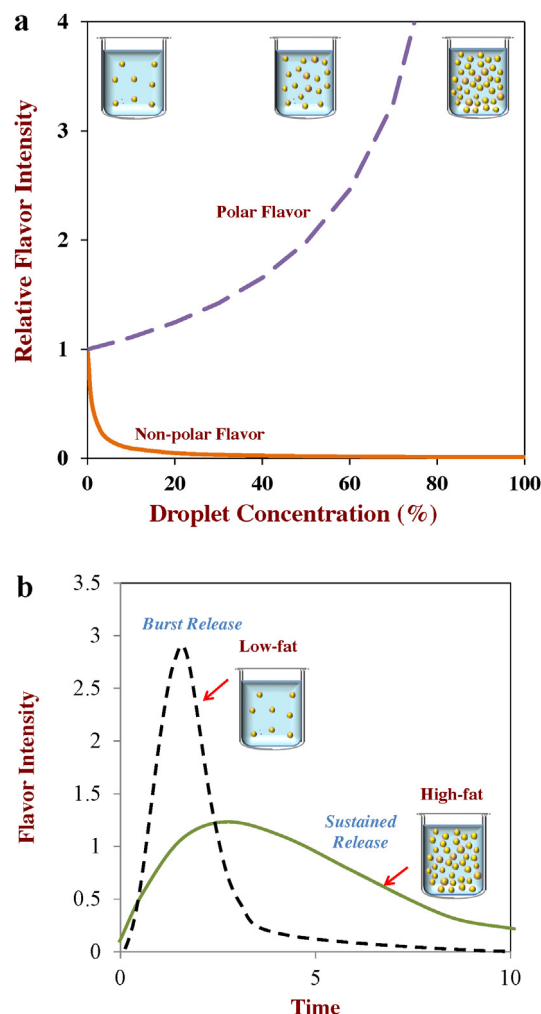


Fig. 6 – (a) Schematic representation of flavor dependence in oil-in-water emulsions on droplet concentration. (b) Schematic diagram of differences in flavor release profiles of low fat and high fat emulsion-based foods. A non-polar flavor molecule will give a “burst release” in a low-fat product, but a more “sustained release” in a high fat product.

phase and headspace above the emulsion (Charles, Rosselin, Beck, Sauvageot, & Guichard, 2000; Karaishou et al., 2008; Miettinen, Tuorila, Piironen, Vehkalahti, & Hyvonen, 2002). In systems containing larger droplets (a few micrometers) there may be a delayed release of the non-polar flavor molecules within the mouth during mastication, as the diffusion path-length increases with droplet size (Charles et al., 2000). Droplet size may also indirectly influence the flavor profile of some food systems through its influence on other physicochemical properties. For example, an investigation of the breakdown of emulsion-filled gels during mastication found that droplet size influence perceived sweetness because it influenced the fracture properties of the gels (Sala & Stieger, 2013).

4.2.2. Continuous phase characteristics

Non-fat particles and other components within the aqueous phase may also influence the perceived flavor characteristics

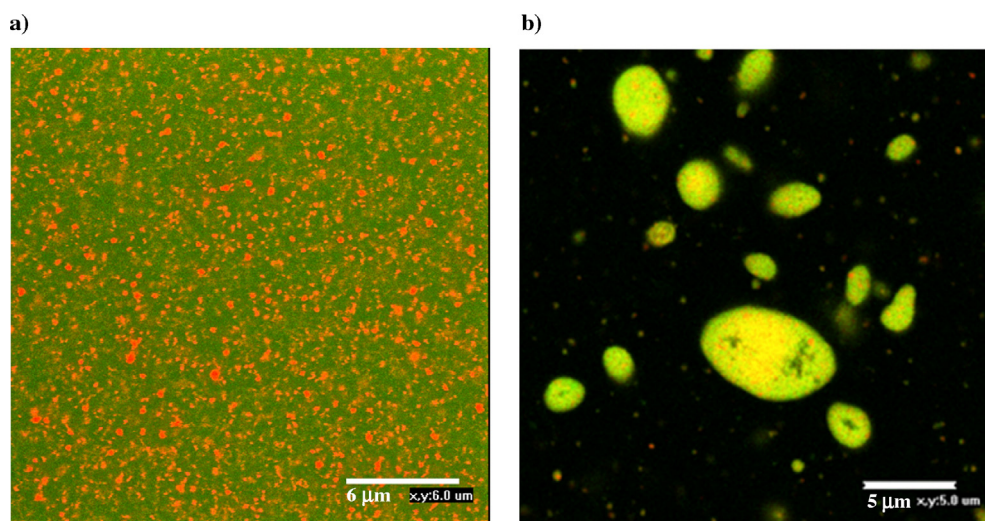


Fig. 7 – Confocal micrographs (60×) of (a) conventional oil-in-water emulsion (1%, w/w oil) stabilized by sodium caseinate (1%, w/w oil) (at pH 7) and (b) oil-filled hydrogel particles (1%, w/w oil) with sodium caseinate as the gel matrix and pectin as the continuous phase at pH 5.

of emulsion-based products due to their ability to alter the partitioning and mass transport of volatile and non-volatile molecules. Thickening or gelling agents can delay diffusion of flavor molecules to the taste receptor on the tongue and hence affect the overall flavor profile (Arancibia, Jublot, Costell, & Bayarri, 2011; Cook, Hollowood, Linforth, & Taylor, 2002; Frank et al., 2012; Gonzalez-Tomas, Carbonell, & Costell, 2004; Hollowood et al., 2008). Some biopolymers alter the perceived flavor of a food by binding flavor molecules (Cook, Linforth, & Taylor, 2003; Rosett, Kendregan, Gao, Schmidt, & Klein, 1996).

4.2.3. Interfacial region

The nature of the interfacial region may also influence the kinetics of flavor release (Charles et al., 2000; Christiansen et al., 2011; Karaishou et al., 2008; Miettinen et al., 2002). Some emulsifiers can bind flavor compounds and delay their release into the mouth and headspace. The nature of the interfacial region also determines how fat droplets behave within the mouth (e.g., their ability to stick to the tongue or to aggregate), which will alter the release of flavor molecules.

4.3. Texture and mouthfeel

The “texture” and “mouthfeel” of emulsion-based food products plays an important role in determining their perceived flavor. Texture is “the sensory and functional manifestation of the structural, mechanical and surface properties of foods detected through senses of vision, hearing, touch and kinesthetic” (Szczeniak, 2002), while “mouthfeel” is generally defined as the sensations arising from the interactions of an ingested food (mixed with saliva) with the receptors in the mouth that respond to tactile stimuli during mastication (Smith & Margolskee, 2001). A range of descriptors is commonly used to describe the texture and mouthfeel characteristics of food emulsions, such as “creaminess”, “richness”, “smoothness”, “sliminess”, “thickness”, “thinness”, “watery”, “firmness”, “hardness”, and “astringency”

(Chen & Eaton, 2012; Guinard & Mazzucchelli, 1996). The influence of the various phases in an emulsion on the texture and mouthfeel of emulsion-based food products is discussed in the following sections.

4.3.1. Dispersed phase characteristics

The type and concentration of fat droplets in an emulsion-based food greatly influences its perceived texture and mouthfeel. Increasing the fat droplet concentration in a product increases the perception of “richness”, “creaminess”, “smoothness”, “thickness”, and “fattiness” (Chojnicka-Paszun et al., 2012; Chung, Olson, et al., 2013; de Wijk & Prinz, 2005; de Wijk & Prinz, 2007; Kilcast & Clegg, 2002; van Aken et al., 2011). The difference between reduced-fat and full-fat versions of food products can be used to demonstrate the influence of fat content on their texture and mouthfeel. In many products, the reduced fat versions are perceived as not being as “rich”, “creamy” or “thick” as their full-fat counterparts, which has been partly attributed to their lower viscosities (Chung et al., 2012b; Chung, Olson, et al., 2013). Some studies have also reported that droplet size influences the perceived “creaminess” of food emulsions (Ciron, Gee, Kelly, & Auty, 2012; de Wijk & Prinz, 2005; Kilcast & Clegg, 2002). de Wijk and Prinz (2005) found that increasing the fat droplet size in mayonnaises increased the measured friction, which is a parameter associated with high roughness and low creaminess. However, other studies reported little influence of fat droplet size on creaminess or thickness (Akhtar et al., 2005; Frost, Dijksterhuis, & Martens, 2001). These differing findings suggest that fats can influence the sensory properties of different food products in distinctive manners.

Droplet interactions: Colloidal interactions between droplets have a significant impact on the rheology and texture of food emulsion products. These interactions affect the effective volume fraction of the dispersed phase in an emulsion, and therefore influence the overall viscosity and texture properties. Attractive interactions between droplets can lead to

droplet flocculation, which increases the effective volume fraction of the dispersed phase due to the presence of the continuous phase trapped within floc structures (McClements, 2012a). Flocculation and the associated increase in viscosity can be induced through several methods, including adjusting solution pH or ionic strength to reduce electrostatic repulsion, addition of biopolymers to increase depletion or bridging attraction, and heating to increase hydrophobic attraction between globular protein-coated droplets (Aben, Holtze, Tadros, & Schurtenberger, 2012; Protonotariou et al., 2013; Tangsuphoom & Coupland, 2008).

Droplet size: The size of the droplets in an emulsion only has an appreciable influence on the viscosity of non-flocculated systems when the dispersed phase volume fraction is relatively high, $\phi > 0.45$ (McClements, 2005). The degree of polydispersity of the oil droplets also influences the viscosity of emulsions since it affects the effective packing parameter. The packing of the droplets in polydisperse emulsions is more efficient than in monodisperse emulsions, since small particles can fit in the spaces between large particles, thereby leading to lower viscosities at similar fat contents (Chung et al., 2012b; McClements, 2005, 2012a).

4.3.2. Continuous phase characteristics

The properties of the aqueous phase may also influence the perceived texture and mouthfeel of food emulsions. The presence of biopolymers, particularly those with thickening or gelling properties, influences the texture and mouthfeel of emulsions (Akhtar, Murray, & Dickinson, 2006; Flett, Duizer, & Goff, 2010; Meyer, Bayarri, Tarrega, & Costell, 2011). The viscosity of emulsions is directly proportional to the viscosity of the continuous phase, and therefore any components within the aqueous phase that enhance its viscosity will influence the overall rheology and textural properties of the system (Cerimedo et al., 2010; Chung et al., 2012b; Huck-Iriart et al., 2011; Perrechil & Cunha, 2010; Protonotariou et al., 2013; Rao, 2007). The degree of the increase depends on the concentration, conformation, and interactions of the components present and the spatial arrangement of the overall system (Fig. 3). Addition of biopolymers can therefore be used as a means of replacing some of the textural attributes lost when fat droplets are removed from reduced-fat products (Chung et al., 2013a; Dickinson, 2013; Flett et al., 2010; Mun et al., 2009; Torres, Janhoj, Mikkelsen, & Ipsen, 2011).

Recently, a novel approach to regulate the rheological properties of emulsion-based products has been developed based on the incorporation of micron-sized air bubbles. Preliminary studies on these air-filled emulsions demonstrated that their rheological properties were comparable to those of full-fat emulsions (Tchuenbou-Magaia & Cox, 2011). A more detailed discussion of the formation and properties of air-filled emulsions is given in Section 6.

4.3.3. Interfacial region

The perceived texture and mouthfeel of emulsions can be altered by modulating the way that the fat droplets behave within the oral cavity, e.g., whether they coalesce and/or spread on the tongue. Studies have reported that fat droplets that are prone to coalescence within the mouth have creamier mouthfeel and fatty sensations than those that do not

(Benjamins et al., 2009; Dresselhuys, de Hoog, Cohen Stuart, Vingerhoeds, & van Aken, 2008a). The attachment of fat droplets to the oral cavity and their tendency to coalesce and spread within the mouth can be controlled by altering the nature of the emulsifier used to stabilize them (Benjamins et al., 2009; Dresselhuys, de Hoog, Cohen Stuart, Vingerhoeds, & van Aken, 2008a; van Aken, 2007).

4.4. Food emulsion stability

Emulsions are thermodynamically unstable systems that are inclined to break down over time through a variety of physicochemical mechanisms, including gravitational separation, flocculation, coalescence and Ostwald ripening (Fig. 8a). Practically, an emulsion-based food product can be considered to be physically unstable when there is visual separation or inhomogeneity in its overall appearance, or an undesirable change in its textural attributes.

Control of emulsion stability is important in the food industry in the creation of commercial products that maintain their desirable sensory and physicochemical properties throughout the products shelf life. There are various possible strategies available to retard emulsion instability by modulating the properties of the dispersed, continuous and interfacial phases.

4.4.1. Disperse phase characteristics

The structural and physicochemical characteristics of the disperse phase affect a number of different emulsion instability mechanisms e.g., gravitational separation, flocculation, coalescence and Ostwald ripening (Fig. 8a). The density of the dispersed phase influences the tendency for gravitational separation to occur. The concentration of droplets in an emulsion also influences their stability to gravitational separation – the higher the droplet concentration, the slower the creaming rate (Fig. 8b). Increasing the droplet concentration may not be a feasible solution to inhibiting gravitational separation in most food products, but it may be possible to introduce other non-fat particles to inhibit creaming by a similar mechanism (e.g., hydrogel particles). The size of the droplets also plays an important role in gravitational separation, with the rate of creaming increasing with the square of the particle radius. Thus, gravitational separation can be retarded by decreasing the droplet size e.g., by altering the homogenization conditions (e.g., increasing the intensity or duration of homogenization) and product formulation (e.g., emulsifier type and concentration) (Cerimedo et al., 2010; Huck-Iriart et al., 2011; Perrechil & Cunha, 2010).

4.4.2. Continuous phase characteristics

The properties of the continuous phase may also influence a variety of different instability mechanisms in emulsions. Addition of substances that increase the viscosity or gel strength of the continuous phase retards droplet movement e.g., starches, gums, sugars, and/or proteins (Cerimedo et al., 2010; Chung et al., 2012b; Huck-Iriart et al., 2011; Perrechil & Cunha, 2010; Protonotariou et al., 2013). Consequently, they may reduce the rates of both droplet gravitational separation and aggregation. Changes in the density of the continuous phase influence the rate of gravitational separation. Alterations

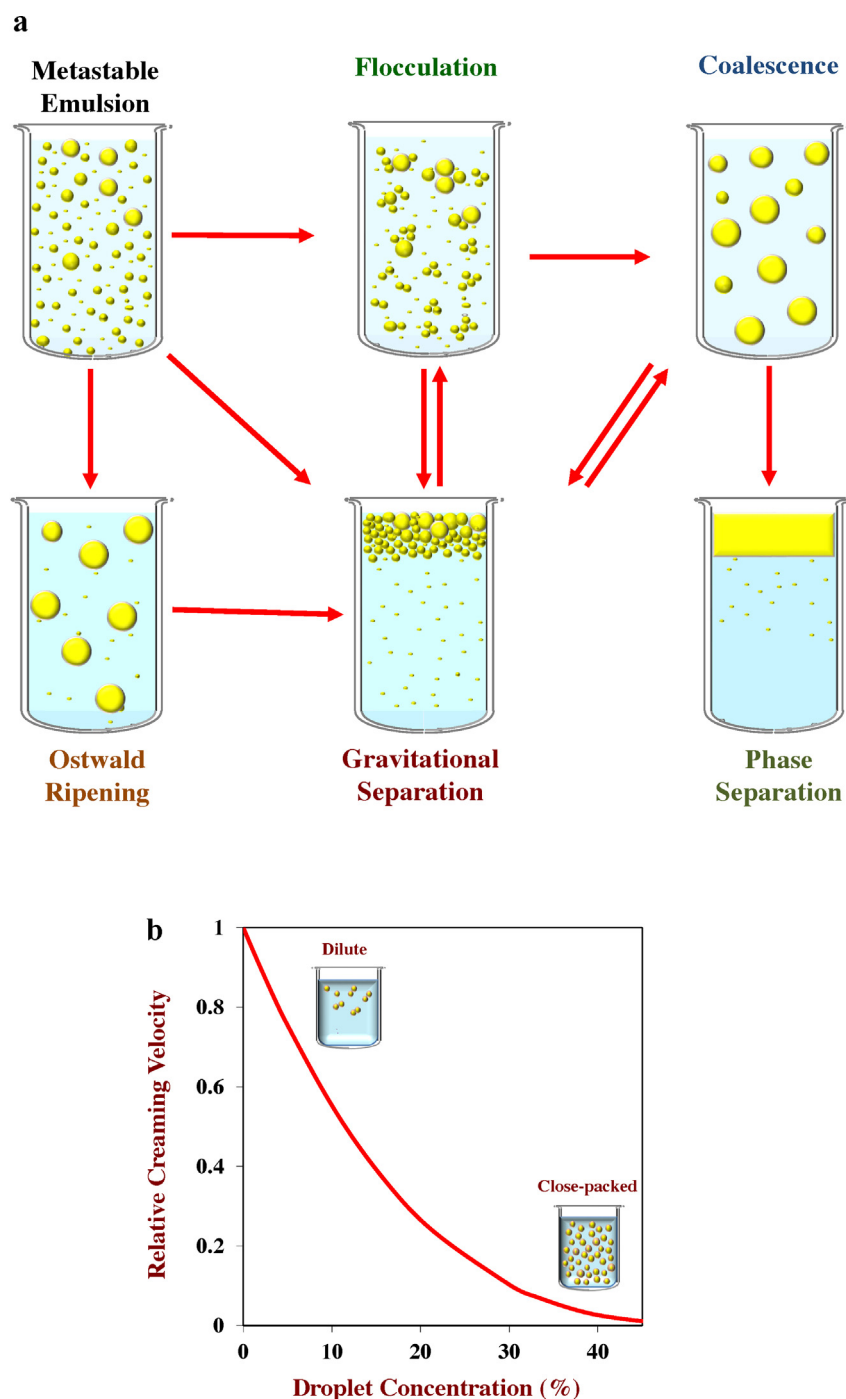


Fig. 8 – (a) Schematic diagram of most common instability mechanisms that occur in food emulsions: flocculation, coalescence, creaming, sedimentation, Ostwald ripening and phase separation. (b) Calculated dependence of the creaming velocity of the oil droplets in oil-in-water emulsions on droplet concentration.

in the pH or ionic strength of the aqueous phase surrounding fat droplets may alter the magnitude and range of colloidal interactions, and therefore their susceptibility to aggregation (Protonotariou et al., 2013; Tangsuphoom & Coupland, 2008). The addition of non-adsorbing biopolymers may induce flocculation by increasing the depletion attraction between droplets (Aben et al., 2012). On the other hand, addition of adsorbing biopolymers may promote flocculation through

bridging mechanisms (Guzey & McClements, 2007; Tran & Rousseau, 2013).

4.4.3. Interfacial region characteristics

The properties of the interfacial region surrounding the oil droplets in an emulsion often play a critical role in determining their stability. One of the most important effects is their impact on the colloidal interactions between droplets, i.e.,

their ability to modulate attractive or repulsion interactions. The thickness of the interfacial layer determines the magnitude and range of the steric repulsion between droplets. Droplet flocculation can be inhibited if the range of the steric repulsion is longer than the range of any attractive interactions, such as van der Waals or hydrophobic (Wooster & Augustin, 2006, 2007). The charge on the interfacial region determines the magnitude of the electrostatic repulsion between oil droplets, and any factors that increase the charge tend to improve stability (Guzey & McClements, 2007; Tang & Shen, 2013; Tran & Rousseau, 2013). The polarity of the interfacial region also influences stability – the more hydrophilic the interfacial layer, the weaker are any hydrophobic attractive forces, and therefore the better the stability to aggregation (Kim, Decker, & McClements, 2002).

5. Instrumental assessment of food emulsion properties

A number of analytical methods have been developed to evaluate the sensory qualities of food products during product development. These instruments are useful for identifying the key factors that influence the sensory attributes of foods, and to provide quantitative information that may predict the sensory performance of foods. However, instrumental methods cannot model the extreme complexity of the human sensory system and hence cannot replace sensory analysis. Nonetheless, both instrumental analyses and sensory evaluations have their advantages and limitations. In this section, we briefly list a range of analytical methods available to measure the different attributes of food, namely appearance, aroma, taste and, texture and mouthfeel, as well as microstructure analyses.

5.1. Appearance

The appearance of food products gives the first impression of their sensory properties and hence it has a significant impact on the subsequent in-mouth perception and acceptability of the food. As mentioned above, the appearance of food emulsions is influenced by the light scattered from the fat droplets, which depends on their concentration and size. The color of food products can be determined using a colorimeter that quantifies the color in tristimulus coordinates *e.g.*, $L^* a^* b^*$ (Gonzalez-Tomas & Costell, 2006; Hutchings, 1999). The L^* represents lightness, where low L^* (0) is dark (black) and high L^* (100) is light (white). The a^* and b^* values are color coordinates, where $+a^*$ is the red direction, $-a^*$ is the green direction and $+b^*$ is the yellow direction, $-b^*$ is the blue direction. The opacity of an emulsion can be characterized by the lightness (L^*), while the color intensity can be characterized by the chroma: $C = (a^{*2} + b^{*2})^{1/2}$. The color intensity is usually inversely related to the lightness, which accounts for the reduction in color intensity when the droplet concentration in emulsions increases (McClements, 2005). Besides color, the surface gloss of food products also has marked influence on consumers' perception and acceptance of the products. Surface gloss is a measure of the light reflection from a food surface whether it is primarily specular (glossy) or diffusive (matte). This

reflection depends on surface characteristics such as topology and heterogeneity. For both color and surface gloss measurements, a variety of handheld and bench top colorimeters are commercially available to measure the color, opacity, and surface gloss of food products conveniently.

5.2. Aroma – volatile compounds

The volatile aromatic compounds released into the headspace of food products and reaching the nasal cavity during mastication can be quantified instrumentally using headspace analysis (Biniecka & Caroli, 2011; Chambers & Koppel, 2013; Dymerski, Chmiel, & Wardencki, 2011; Frank et al., 2011; Rabe, Krings, Banavara, & Berger, 2002; Taylor & Linforth, 2000). Static headspace analysis is employed to determine the equilibrium partition coefficient of flavor compounds between a food and the headspace. Dynamic headspace analysis is used to measure flavor release rates, and may be carried out on the headspace collected above a food product or from the breath exhaled from a human during mastication. The composition of the flavor compounds collected in the headspace can be determined using several methods, such as gas chromatography (GC), high performance liquid chromatography (HPLC) and mass spectrometry (Biniecka & Caroli, 2011; Dymerski et al., 2011; McClements, 2005; Taylor & Linforth, 2000). Electronic noses have also been developed to simulate the response of human sensory receptors to food flavor and have been useful for monitoring changes in the concentrations of flavor compounds over time above a product (Biniecka & Caroli, 2011; Dymerski et al., 2011; Ravi, Prakash, & Bhat, 2013; Zawirska-Wojtasiak & Mildner-Szkudlarz, 2010).

5.3. Taste – non-volatile compounds

Some non-volatile compounds can elicit a taste response during mastication *e.g.*, salty, sweet, sour, bitter, or umami. Taste is normally attributed to non-volatile compounds soluble in the aqueous phase. Some taste molecules may partition between the different phases within an emulsion, which alters their perception by reducing the amount in the aqueous phase. The characterization of taste typically involves measurements of the time-dependence of taste molecule concentrations in the aqueous phase, which is influenced by their oil–water partition coefficients and oil-to-water mass transfer rates. Static and dynamic methods can be used to measure the concentration of non-volatile compounds in the water phase within a food product or within the mouth during mastication by collecting and analyzing saliva samples over time (Calvino, Garrido, & Garcia, 2000; Sokolowsky & Fischer, 2012). In the last decades, electronic tongues have been developed to assess and quantify the intensity of tastes in foods and beverages (Eckert, Pein, Reimann, & Breitzkreutz, 2013; Lorenz, Reo, Hendl, Worthington, & Petrossian, 2009; Rudnitskaya et al., 2013; Tahara & Toko, 2013). Sophisticated devices can be designed by coupling electronic tongues and noses in the same instrument (Baldwin, Bai, Plotto, & Dea, 2011; Cole, Covington, & Gardner, 2011; Kovacs et al., 2011). Electronic tongues are designed to mimic the human taste sensors and their communication with the brain.

5.4. Texture and mouthfeel

The most commonly used instrumental methods to quantify perceived textural attributes (e.g., thickness, creaminess, smoothness) of food products are based on rheological measurements, such as viscosity, viscoelasticity, and fracture properties (Fischer, Pollard, Erni, Marti, & Padar, 2009; Fischer & Windhab, 2011; McClements, 2005; Stokes, Boehm, & Baier, 2013; van Aken, 2007). However, these measurements carried out using a viscometer or rheometer may not closely simulate the complex processes that occur during mastication within the mouth, including product fracture, interactions with saliva, coating of the tongue and palate, and complicated stress/flow profiles (de Wijk, Janssen, & Prinz, 2011; Foegeding et al., 2011; Stokes et al., 2013; Bilt, Engelen, Pereira, van der Glas, & Abbink, 2006). Nevertheless, close correlations found between rheological measurements and sensory perception have been reported for many products (Akhtar et al., 2006; Chung, Olson, et al., 2013; Melito, Daubert, & Foegeding, 2013; Mossaz et al., 2010; Vidigal et al., 2012). Alternative instrumental methods that closely mimic in-mouth mastication have been developed and utilized in the recent decades, as will be discussed in the following sections.

5.4.1. Large deformation flow behavior

Large deformation measurements are particularly useful for monitoring the rheology and disruption of emulsion-based products (Foegeding et al., 2011). They can be applied in shear, compression, or combined modes. The apparent viscosity of samples, calculated as the shear stress divided by the rate of strain, is commonly used to indicate the “thickness” of liquid to semi-solid food emulsion products. Food emulsion products often exhibit non-Newtonian behavior, i.e., their viscosity is dependent on the rate of strain (Chung et al., 2013c; Gonzalez-Tomas, Bayarri, Taylor, & Costell, 2008; McClements, 2005; Ochowiak, Broniarz-Press, & Rozanski, 2012). It is important to select strain rates that simulate the flow conditions of food emulsions during mastication. It is also essential to mimic other conditions within the mouth such as dilution with saliva and mechanical disruption during mastication. This is particularly significant in foods that contain digestible starch because starch granules are degraded by amylase during mastication (Chung et al., 2012a; Chung, Olson, et al., 2013). Many emulsion-based food products have plastic-like characteristics i.e., below a critical applied stress (the yield stress) no flow is measured and above it samples behave as viscous fluids. These food products are usually characterized in terms of their yield stress, consistency, and flow index (McClements, 2005).

5.4.2. Small deformation viscoelastic properties

For gel and solid food products the most commonly measured property is the dynamic shear modulus (G), which has as an elastic component G' (the storage modulus) and a viscous component G'' (the loss modulus). The dynamic shear modulus can also be reported as a complex modulus (G^*) and a phase angle (δ) (Dogan, Kayacier, Toker, Yilmaz, & Karaman, 2013; Dolz, Hernandez, & Delegido, 2006; Liu & Tang, 2011; Mun et al., 2009). Again, these measurements can be made in shear, compression or combined modes. It is also important to establish the most appropriate operating conditions for small

deformation measurements, e.g., the oscillation frequency and strain. A frequency of 50 rad s^{-1} has been reported to give a good correlation with the perceived “thickness” of some foods (Bistany & Kokini, 1983; Richardson, Morris, Ross-Murphy, Taylor, & Dea, 1989; Stokes et al., 2013). It should be stressed that small deformation measurements are carried out under very different conditions than those within the mouth during mastication.

5.4.3. Tribology

Alternative instrumental methods that more accurately model the complex processes occurring within the mouth during mastication have been developed and utilized in recent years. For example, a thin lubricating film of food is formed between the tongue and palate during mastication and hence it is an important element that should be simulated during instrumental analysis. Thin film rheology (“tribology”) has increasingly been used to simulate the oral behavior of food products and to correlate to sensory perception (Chen & Stokes, 2012; Dresselhuys, de Hoog, Stuart, & van Aken, 2008b; Krzeminski, Wohlhuter, Heyer, Utz, & Hinrichs, 2012; Stokes et al., 2013). In this method, an instrument known as a “tribometer” is used to measure thin film rheology. A simple tribometer consists of a ball and a disk with a sample placed between them. During measurements a normal load is applied to the system and the ball and disk rotate at different speeds leading to a relative strain rate between them. The instrument then measures the coefficient of friction of the material as the strain rate is varied. Studies have shown that tribology measurements can provide useful information about food products that cannot be obtained from conventional bulk rheology methods. The study of tribology (friction, lubrication and adhesion) on some food products has also been related to in-mouth textural perception e.g., creaminess (Chen & Stokes, 2012; Dresselhuys, de Hoog, Stuart, & van Aken, 2007; Krzeminski et al., 2012; Reverend, Norton, Cox, & Spyropoulos, 2010; Stokes et al., 2013).

5.4.4. Extensional/elongational flow rheology

Elongational flow has also been recognized to play an important role during the mastication of food products (de Bruijne, Hendrickx, Anderliesten, & de Looff, 1993). This is because when the tongue and palate move together during mastication the food material is squeezed out from between them, which produces a strong elongational flow component. Thus, an ideal instrumental method to determine and quantify textural properties should include both elongational and shear flow to more accurately simulate oral food texture (Le Reverend et al., 2010; Lillford, 2000). A number of testing methods have been developed that utilize elongational and shear flow, such as tribology (see above), squeezing flow (Campanella & Peleg, 2002; Chatraei, Macosko, & Winter, 1981; Corradini & Peleg, 2005; Pelot, Sahu, Sinha-Ray, & Yarin, 2013), “imperfect” squeezing flow (Damrau & Peleg, 1997; Hoffner, Gerhards, & Peleg, 1997; Terpstra, Janssen, & van der Linden, 2007), and pressure drop techniques (Carrier et al., 2013; Macosko, 1994).

Recently, a combined squeezing flow and shear viscosity method was developed to simulate the oral processes occurring during mastication (Chung, Degner, & McClements,

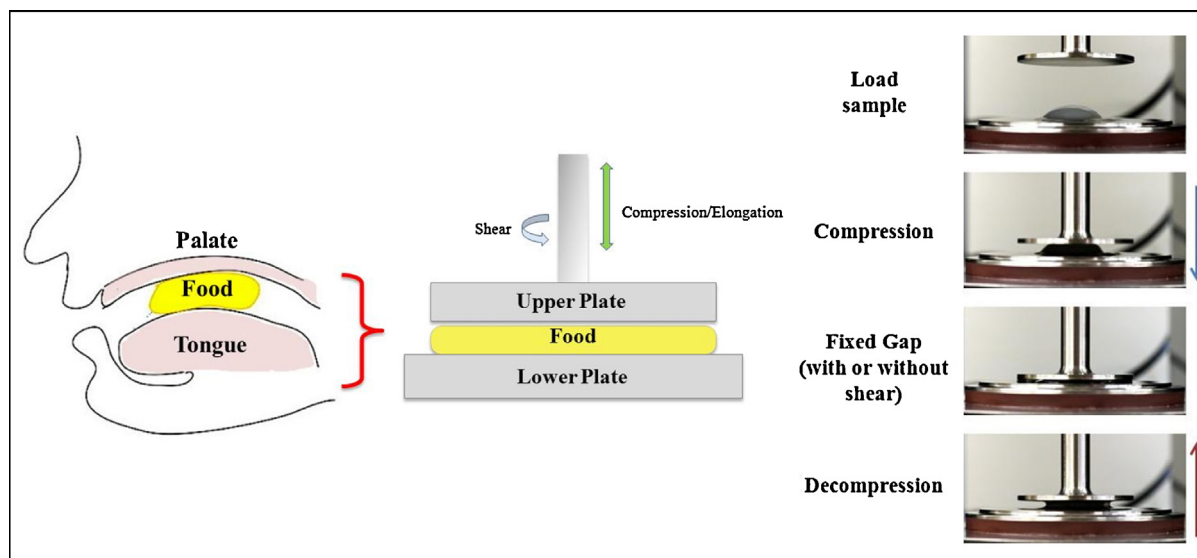


Fig. 9 – Schematic representations and photographic images of the instrumental mastication device designed to simulate the tongue and palate in the mouth.

2012a; Chung, Olson, et al., 2013). This method utilizes two horizontal parallel plates of a rheometer with the sample to be tested placed between them. The sample then undergoes a series of sequential compression-shear-decompression motions to simulate in-mouth mastication (Fig. 9). The normal and shear forces are measured on the upper plate during measurements, which provide information about the rheological properties of the sample, such as maximum peak force (related to consistency), maximum trough force (related to adhesiveness), residual stress (related to yield stress), and shear viscosity. Also, saliva can be added to the sample prior to the start of measurement to more closely simulate oral conditions (Chung et al., 2012a; Chung, Olson, et al., 2013).

5.5. Microstructure analysis

The spatial arrangement of the various components within a food emulsion plays a major role in determining its physico-chemical and sensory properties, *e.g.*, droplet size, location, and interactions. Consequently, it is important to have appropriate analytical tools to determine emulsion microstructure. This knowledge may then shed light on the mechanisms responsible for overall system properties. The microstructure of emulsions can be characterizing using various microscopy instruments *e.g.*, optical microscopy, confocal fluorescence laser scanning microscope, scanning electron microscope, and transmission electron miscopy (McClements, 2005). Some examples of the microstructures of different emulsion-based food systems obtained using different microscopy methods are shown in this review (*e.g.*, Figs. 3, 5, 7 and 11).

6. Structural design approaches

Recently, there has been great interest in extending the functional performance of food emulsions using various

structural design approaches. In this section, a brief introduction to some of these structural design approaches is provided, and potential applications within the food industry are highlighted.

6.1. Multiple emulsions

A mixture of water-in-oil and oil-in-water elements contained within the same system is known as a multiple emulsion. Structurally, multiple emulsions consist of small droplets of one phase embedded within larger droplets of a second phase, which itself is dispersed within a continuous phase (Figs. 1 and 10). The two major types of multiple emulsions are water-in-oil-in-water ($W_1/O/W_2$) and oil-in-water-in-oil ($O_1/W/O_2$) emulsions (Dickinson, 2011; Leal-Calderon et al., 2007; McClements, 2012a; Muschliolik, 2007). The subscripts 1 and 2 refer to the inner and outer regions of one of the liquid phases (either oil or water). For example, for $W_1/O/W_2$ emulsions the W_1 indicates the inner aqueous phase and the W_2 indicates to the outer aqueous phase. These two aqueous phases may have different compositions and properties (McClements, 2012a). $W_1/O/W_2$ emulsions are suitable for various applications within aqueous-based foods since they can be used to produce reduced-fat products and for encapsulation and controlled release of bioactive molecules (Dickinson, 2011; Leal-Calderon et al., 2007).

Multiple emulsions can be prepared using many of the same tools used for preparation of conventional emulsions, *e.g.*, high shear mixers, high pressure homogenizers, sonicators, and membrane homogenizers (McClements, 2012a; Muschliolik, 2007). $W_1/O/W_2$ emulsions are normally produced using a two-stage process. In brief, the first stage involves formation of a W_1/O emulsion by homogenizing water and oil together in the presence of an oil-soluble surfactant. The second stage involves homogenization of the W_1/O emulsion with water in the presence of a water-soluble surfactant to

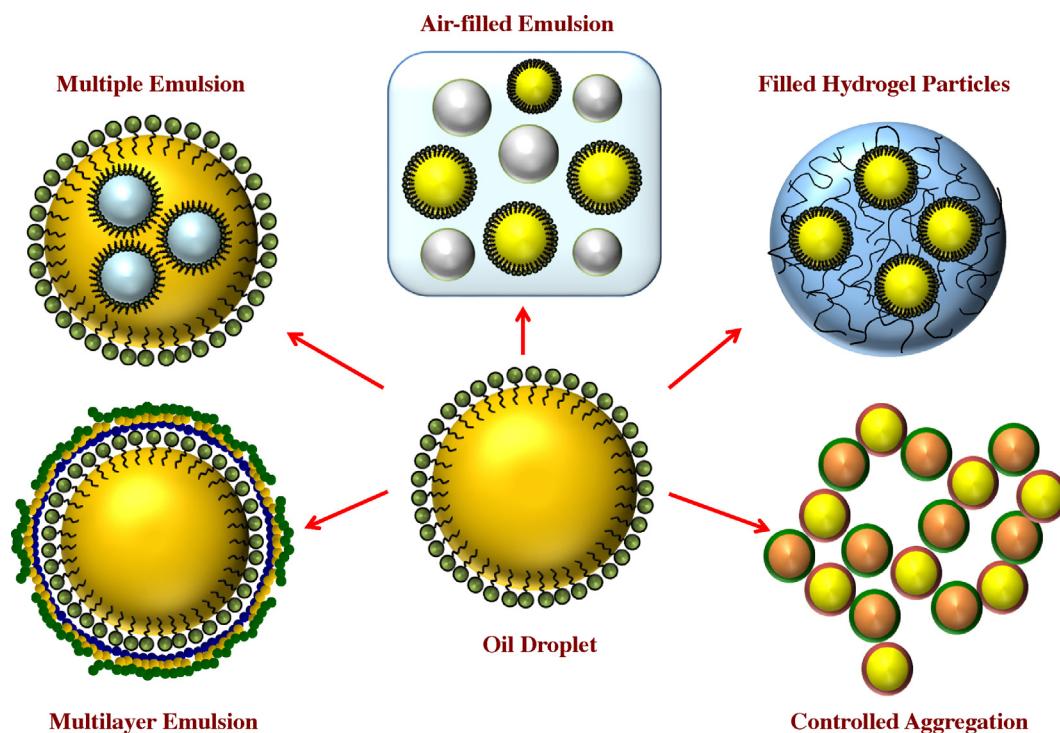


Fig. 10 – Examples of structured emulsions created by structural design principles using emulsion droplets as a building block.

form a $W_1/O/W_2$ emulsion (Dickinson, 2011; McClements, 2012a). Usually, the second homogenization stage has to be less intense than the first otherwise the system may breakdown (McClements, 2012a; Pal, 2011). Multiple emulsions are thermodynamically unstable systems that break down over time and therefore they must be carefully designed to give them sufficient kinetic stability for practical applications. $W_1/O/W_2$ emulsions breakdown due to the same mechanisms as conventional emulsions (such as gravitational separation, droplet aggregation, and Ostwald ripening), plus some additional mechanism (such as water diffusion and droplet expulsion from the oil droplets) (Dickinson, 2011; McClements, 2012a). Further details of the formation and instability of multiple emulsions are available from the cited references.

A number of studies have investigated the potential application of multiple emulsions ($W_1/O/W_2$) in food products (Giroux et al., 2013; Lobato-Calleros et al., 2008; O'Dwyer, O'Beirne, Ni Eidhin, Hennessy, & O'Kennedy, 2013). For example, it was shown that reduced fat cheeses could be prepared that had properties similar to full fat cheeses (Lobato-Calleros, Rodriguez, Sandoval-Castilla, Vernon-Carter, & Alvarez-Ramirez, 2006; Lobato-Calleros et al., 2008).

6.2. Filled hydrogel particles

Filled hydrogel particles are systems in which the oil droplets are embedded within a hydrogel matrix that is itself dispersed within an aqueous continuous phase (Figs. 7 and 10). The hydrogel particles can be made from proteins and/or polysaccharides that are capable of forming a gel matrix under

controlled conditions (e.g., heating, cooling, pH adjustment, or enzyme addition). Filled hydrogel particles can be formed using a variety of methods, including injection methods, molding methods, gel disruption, and controlled biopolymer phase separation (Burey, Bhandari, Howes, & Gidley, 2008; Matalanis, Jones, & McClements, 2011; McClements, 2012a). Most of these methods involve mixing an O/W emulsion with a biopolymer solution and then adjusting the preparation conditions to promote hydrogel particle formation and matrix gelation (McClements, 2012a).

Hydrogel particles are often formed using controlled biopolymer phase separation, which can be divided into segregation-based and aggregation-based methods (Schmitt & Turgeon, 2011). Segregation-based separation occurs due to thermodynamic incompatibility of the two biopolymers (Norton & Frith, 2001). Thermodynamic incompatibility arises when there is a repulsive interaction between two different kinds of biopolymers. Above a critical concentration, the mixed systems separate into two biopolymer phases with different compositions, one phase that is rich in one type of biopolymer and depleted in the other, and vice versa (Norton & Frith, 2001; Norton & Norton, 2010). Hydrogel particles are then formed by blending the system or using injection methods. The hydrogel particles are then cross-linked using a method that depends on biopolymer type, such as adjustment of temperature, pH, salt, or enzyme activity. The preparation method is designed so that the oil droplets are located within the biopolymer phase that forms the hydrogel particles. The viscosity of filled hydrogel particles is higher than that of conventional emulsions with the same oil concentration due

to the increased effective volume fraction of the hydrogel matrices (Matalanis et al., 2011). Filled hydrogel particles are thus suitable for application in reduced fat products, which need to attain high viscosity when fats are removed. Filled hydrogel particles may also be used to manipulate the release of flavor compounds from the oil phase (Malone & Appelqvist, 2003; Raschip, Hitruc, Oprea, Popescu, & Vasile, 2011). With the oil trapped in the hydrogel matrices, there is an increased path-length through which the flavor compounds need to diffuse before they can be released from the systems during mastication. Thus, the flavor release profile of a reduced-fat product may be matched to that of the full-fat counterpart (Fig. 6b).

6.3. Multilayer emulsions

The properties of O/W emulsions can also be modified by building laminated coatings around the oil droplets using the layer-by-layer (LbL) deposition method (McClements, 2010a). The oil droplets can be surrounded by a nano-laminated shell consisted of the emulsifier interfacial layer and one or more polyelectrolyte coatings around the adsorbed emulsifier (Fig. 10). The characteristics of the layers (e.g., charge, thickness, integrity, and permeability) can be tailored for specific functional performances (e.g., improved stability or controlled release) by choosing appropriate emulsifiers, polyelectrolytes, and preparation conditions (Guzey & McClements, 2006; McClements, 2012a; Shchukina & Shchukin, 2012).

In brief, the preparation of multilayer emulsions involves mixing an O/W emulsion stabilized by a charged emulsifier with an aqueous solution containing an oppositely charged polyelectrolyte under conditions where the polyelectrolyte forms a coating around the droplet surfaces. Additional layers can be added by mixing the coated droplets with more oppositely charged polyelectrolyte solutions (Guzey & McClements, 2006; McClements, 2012a; Shchukina & Shchukin, 2012). More detailed information on fabrication of multilayer emulsions can be found in the cited references. The formation of multilayer emulsions must be carefully controlled to avoid bridging flocculation, e.g., by controlling oil content, polyelectrolyte concentration, pH, and ionic strength (Guzey & McClements, 2006; Shchukina & Shchukin, 2012).

6.4. Microclusters

The viscosity of emulsions can be increased considerably by inducing droplet flocculation, which increases the effective volume of the particles due to the aqueous phase trapped within the flocs (Fig. 10) (Dickinson, 2009a; McClements, 2005). At sufficiently high droplet concentrations, flocculation can form a three-dimensional network that results in gel-like or paste-like characteristics in a product and inhibits gravitational separation (McClements, 2005). Controlled flocculation is useful in reduced fat foods where a high viscosity needs to be attained after fat droplets are removed (Mao & McClements, 2011, 2012). The formation of microclusters with specific properties can be achieved by homoaggregation or heteroaggregation. Homoaggregation involves aggregation of similar particles, whereas heteroaggregation involves aggregation of dissimilar particles.

Homoaggregation can be induced in food emulsions using various approaches: (i) reduction of electrostatic repulsion between fat droplets by altering the pH to reduce the charge on the droplets or by adding salts to screen the charges (Dickinson, 2010; McClements, 2012a); (ii) increase depletion attraction – by adding non-adsorbing polymers or colloidal particles to increase the osmotic pressure and generate a depletion attraction between the droplets (Dickinson, 2010; McClements, 2012a; Udomrati, Ikeda, & Gohtani, 2011); (iii) increase hydrophobic attraction in globular-protein-stabilized droplets by heating the emulsions above the thermal denaturation temperature of the adsorbed proteins, which increases the surface hydrophobicity (Dickinson, 2010; Kim et al., 2002); (iv) bridging flocculation – by adding oppositely charged polyelectrolytes or biopolymers to form ionic bridges between different droplets (Cho & McClements, 2009; Dickinson, 2010; Guzey & McClements, 2007).

Heteroaggregation can be achieved by mixing two emulsions together containing oppositely charged fat droplets (Mao & McClements, 2011). This has been achieved by mixing positive droplets (protein-coated) with negative droplets (modified starch-coated) under acidic conditions, or by mixing two protein-coated droplets with different isoelectric points at an intermediate pH where they have opposite charges (β -lactoglobulin and lactoferrin) (Fig. 11). The mixed emulsions produced have much higher viscosities than the individual non-flocculated emulsions they were prepared from. The functional properties of the flocculated emulsions depend on the nature of the flocs formed, such as their concentration, size, shape, internal structure, and bond strength. Nevertheless, controlled flocculation should be carried out with care so as to not cause adverse effects, such as decreased creaming stability.

6.5. Air-filled emulsions

Air-filled O/W emulsions have recently been investigated as a novel approach for producing food products with novel properties or reduced calorie contents (Dickinson, 2013; Le Reverend et al., 2010; Tchuembou-Magaia, Al-Rifai, Mohamed-Ishak, Norton, & Cox, 2011; Tchuembou-Magaia & Cox, 2011; Tchuembou-Magaia, Norton, & Cox, 2009, 2010). In this approach, micron-sized air bubbles are incorporated into O/W emulsions to form triphasic air-in-oil-in-water emulsions, where a significant amount of the fat phase is replaced by air bubbles (Fig. 10). Studies with model salad dressings have shown that triphasic emulsions can give rheological (Tchuembou-Magaia et al., 2009) and tribological (Tchuembou-Magaia & Cox, 2011) properties similar to those of conventional O/W emulsions.

Initially, micron sized air bubbles were stabilized using a novel group of proteins (hydrophobins) that assemble at the air/water interface using a sonication process (Tchuembou-Magaia et al., 2009, 2010, 2011). The interfacial coating formed around the air bubbles by these proteins was shown to be relatively robust and capable of surviving various processing steps during food production. The air-filled emulsions also showed little change in air bubble size when stored for up to 4 months at room temperature (Tchuembou-Magaia et al., 2009). However, hydrophobins are relatively expensive ingredients

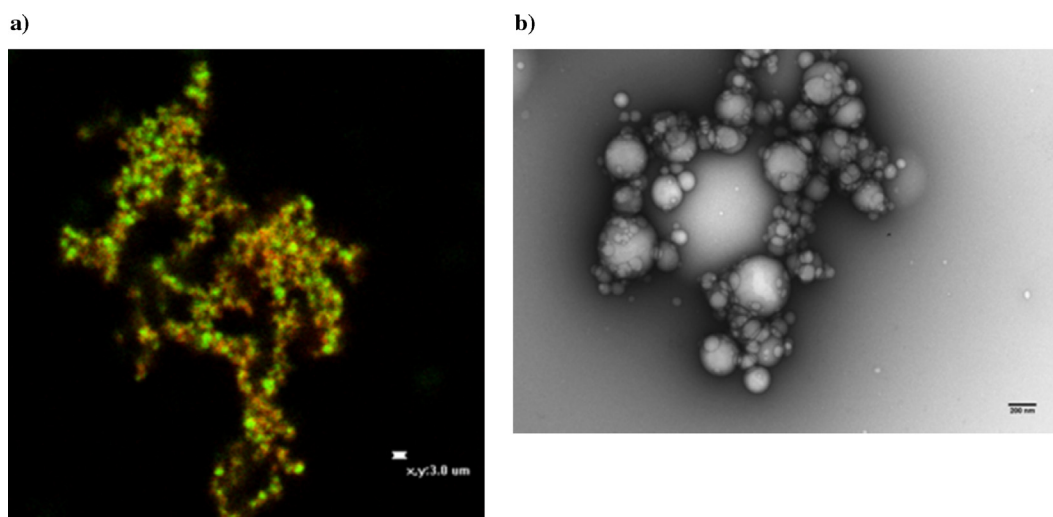


Fig. 11 – (a) Confocal micrograph and (b) transmission electron micrograph of heteroaggregates of lactoferrin-stabilized oil droplets and β -lactoglobulin-stabilized oil droplets. Micrographs were contributed by Mao, Y.Y. (Food Science Dept, UMass, Amherst).

that are not currently suitable for widespread application within the food industry, and so alternative sources of proteins are being examined (Le Reverend et al., 2010). Recently, it has been shown that cysteine-rich proteins (such as bovine serum albumin and egg white proteins) can also be used to form stable air bubbles using a sonochemical method (Tchuenbou-Magaia & Cox, 2011). The majority of the air bubbles formed by these proteins had diameters between 0.5 and 10 μm . This study also found that the air bubble size and stability is highly dependent on the processing conditions and system composition. These preliminary studies suggest that air-filled emulsions have potential application in manufacturing reduced- or low-fat products with comparable sensory properties as full-fat versions.

7. Conclusions

Oil-in-water emulsions serve as one of the few examples where basic physical and chemical principles can be applied to understand the properties of complex food systems. Most emulsion-based food products are compositionally and structurally complex materials and it is important to appreciate this complexity when designing and fabricating food emulsions. In this article, we have provided a brief overview of the relationship between the structural properties of emulsions and their bulk physicochemical and sensory properties. This knowledge is useful for the rational design of food products with novel or improved functional performances.

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