



Review

Re-coalescence of emulsion droplets during high-energy emulsification

Seid Mahdi Jafari^{a,*}, Elham Assadpoor^a, Yinghe He^b, Bhesh Bhandari^c^a*Department of Food Science and Technology, Gorgan University of Agricultural Sciences and Natural Resources, Gorgan, Iran*^b*School of Engineering, James Cook University, Townsville, Australia*^c*School of Land and Food Sciences, University of Queensland, Brisbane, Australia*

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Abstract

Emulsion droplet size plays a key role in many emulsion properties such as stability, color, appearance, texture, and rheology. Accordingly, different emulsions have been classified based on emulsion size including microemulsions and submicron (nano) emulsions. The aim of emulsification is usually to produce emulsion droplets as small as possible, and various emulsification techniques can be used in this regard. One of the main problems of producing very fine emulsions with high-energy emulsification techniques such as microfluidization is the occurrence of “over-processing”, which is increase in emulsion size by supplying more energy due to a high rate of re-coalescence of new droplets. Different reasons have been mentioned for “over-processing”, including low adsorption rate of the surface-active agent, low residence time of the emulsion in the emulsification zone, high rate of coalescence frequency, and extreme amount of energy density. This review highlights re-coalescence of new droplets during high-energy emulsification along with some common and important emulsification techniques and different factors affecting emulsion droplet size, and thereby re-coalescence.

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*Corresponding author. Tel./fax: +98 171 4426432.

E-mail address: smjafari@gau.ac.ir (S.M. Jafari).

1. Introduction

An emulsion can be simply defined as “a system comprised of two immiscible liquids, one of which is dispersed as droplets (the dispersed or internal phase) throughout the other (the continuous or external phase)” (Becher, 2001; Friberg & Larsson, 1997). Depending on the emulsion droplet size¹ (EDS), emulsions can be divided into micro- (10–100 nm), mini (nano)- (100–1000 nm), and macro-emulsions (0.5–100 μm) (Windhab, Dressler, Feigl, Fischer, & Megias-Alguacil, 2005). Some of the similarities and differences between these emulsions are presented in Table 1.

Many emulsion properties such as stability, rheology, appearance, color, texture, and shelf-life depend on EDS and size distributions (Becher, 2001; McClements, 2005). Accordingly, it has been well documented that EDS plays an important role in the retention of volatiles and surface oil content of the powders during microencapsulation by spray drying (Liu et al., 2001; Liu, Furuta, Yoshii, & Linko, 2000; Risch & Reineccius, 1988; Soottitantawat et al., 2005; Soottitantawat, Yoshii, Furuta, Ohkawara, & Linko, 2003). So, information about EDS is of considerable importance and it is vital to control, predict, measure, and report the size of the droplets in emulsions prepared for different purposes.

The production and control of submicron emulsions with a narrow size distribution have been attracting considerable attention in food, pharmaceutical, and cosmetic industries in recent years. To produce these emulsions, either a large amount of energy or surfactant or the combination of both is required. “Low-energy emulsification” methods like the phase inversion temperature technique involve transitional inversion induced by changing factors that affect the HLB of the system, such as temperature, electrolyte concentration, etc., or catastrophic inversion induced by increasing the dispersed phase volume fraction (Forgiarini, Esquena, Gonzalez, & Solans, 2001; Izquierdo et al., 2002; Tadros, Izquierdo, Esquena, & Solans, 2004). These methods have several limitations such as requiring a large amount of surfactants and a careful selection of surfactant–cosurfactant combination, and are not applicable to large-scale industrial productions (Seekkuarachchi, Tanaka, & Kumazawa, 2006).

On the other hand, “high-energy emulsification” methods, which are the scope of this work, are applicable to industrial operations because of flexible control of EDS distributions, and the ability to produce fine emulsions from a large variety of materials. The energy needed for the production of such fine emulsions can generally be achieved in high-pressure systems, with emulsification pressures of up to 350 MPa (Floury, Desrumaux, Axelos, & Legrand, 2003; Floury, Legrand, & Desrumaux, 2004)

and even up to 700 MPa (Perrier-Cornet, Marie, & Gervais, 2005), nowadays EDS of less than 0.1 μm are achievable. The most commonly used high-energy emulsification techniques are rotor–stator and high-pressure systems that will be reviewed in this section. Stang, Schuchmann, and Schubert (2001), Schultz, Wagner, Urban, and Ulrich (2004), and recently Urban, Wagner, Schaffner, Roglin, and Ulrich (2006), and Seekkuarachchi et al. (2006) provide some good overviews of high-energy emulsification techniques.

2. Techniques of emulsification

The emulsification technique can be simply a form of mixing. In fact, (pre) emulsions are prepared mostly by mixing equipment (Becher, 2001). Modern emulsions, however, can be produced by specially designed devices including high-pressure, ultrasonic, rotor–stator, and membrane systems (Schultz et al., 2004; Urban et al., 2006). In laboratory studies and most emulsion preparations, it is more efficient and convenient to produce an emulsion in two steps: (a) conversion of separate oil and water phases into a “coarse emulsion” with fairly large EDS (usually by rotor–stator devices) and then (b) final reduction of EDS using another technique (e.g. high-pressure systems).

2.1. Rotor–stator systems

These systems are widely used to emulsify liquids with medium to high viscosity (McClements, 2005). For discontinuous operation, agitators or gear-rim dispersion machines are usually used, while for continuous operation, colloid mills with smooth or toothed rotors and stators are available (Urban et al., 2006). The liquid is fed into the colloid mill in the form of a coarse emulsion (Maa & Hsu, 1996a, 1996b; Pinnamaneni, Das, & Das, 2003), or as separate phases, and flows through a narrow gap between a rotating disk (rotor) and a static disk (stator). The rotor/stator assembly consists of a rotor housed concentrically inside the stator with two or more blades and a stator with either vertical or slant slots. As the rotor rotates, it generates a lower pressure to draw the liquid in and out of the assembly, thereby resulting in circulation and emulsification (Maa & Hsu, 1996a, 1996b).

One of the two major forces that can reduce the EDS is mechanical impingement against the wall due to high fluid acceleration. Another force is the shear stress in the gap between rotor and stator, which is generated by the rapid rotation of the rotor. The intensity of the shear stress can be altered by varying the thickness of the gap (about 50–1000 μm), varying the rotation speed (about 1000–25,000 rpm), or by using disks that have toothed surfaces or interlocking teeth (Becher, 2001; McClements, 2005). Obviously, emulsification intensity (power) and the residence time that emulsion droplets stay in the shearing field are the primary parameters to control EDS. Other

¹In rest of the discussion, instead of using different terms such as mean droplet diameter, droplet size, emulsion size, etc., which may become confusing, emulsion droplet size or simply EDS will be used.