Preparation and Characterization of Micro/Nano-emulsions Containing Functional Food Components*

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Emulsions are dispersions of at least two immiscible liquids, one of which is dispersed as droplets in the other liquid, and stabilized by an emulsifier, such as oil and water. Different types of emulsions can be formulated according to different applications. Emulsions are categorized as simple or multiple type. Oil-in-water (O/W) and water-in-oil (W/O) are the simple emulsions, while water-in-oil-in-water (W/O/W), and oil-in-water-in-oil (O/W/O) emulsions are known as multiple emulsions. Droplets of different sizes and the size distribution patterns formed based on different emulsification processes can affect their physicochemical properties. Hence, the droplet size and its distribution may determine the shelf-life stability, rheological properties, color and taste of food emulsions. Micro/nano-emulsions have been increasingly utilized in the food industry as delivery system carriers that can encapsulate, protect, and deliver lipophilic functional food components, such as bioactive lipids, oil-soluble flavors, vitamins, and nutraceuticals. The application of micro/nano-emulsions has potential advantages in increasing the bioavailability of lipophilic functional food components, modulating the product texture and improving the stability of droplets against aggregation. This article provides an overview of the current status of micro/nano-emulsion containing functional food components, preparations and characterizations, and presents the applications of micro/nano-emulsions in food industry.

Keywords: emulsification, micro/nano-emulsion, functional food components, micro/nano device, delivery system

1. Introduction

Emulsion can be defined as two immiscible liquids such as oil and water, with one of the liquids dispersed as small droplets in another. One of the immiscible liquids, defined as the disperse phase, is dispersed as small droplets in another liquid, defined as the continuous phase. To form a stable emulsion, a substance with emulsifying property has to be included, such substances are defined as emulsifiers [1]. As the same composition of emulsion, the droplets size and droplet size distribution greatly affect the properties of the emulsion. The stability of emulsions can be improved with small droplet sizes and narrow size distributions. A conventional emulsion typically has a droplet diameter between 0.01 to 100 μm . It is thermodynamically unstable due to the relatively high interfacial tension associated with the contact between the oil and water phases. A nano-emulsion generally has mean droplet diameter between 20 to 200 nm [2-3].

Emulsion types can be classified into different groups, such as oil-in-water (O/W), water-in-oil (W/O), and multiple emulsions. O/W emulsions consist of oil droplets in a continuous water phase, and W/O emulsions consist of water droplets in a continuous oil phase. Multiple emulsions are more complex systems, and consist of smaller droplets dispersed in larger droplets that are dispersed in a continuous phase. For instance, water-in-oil-in-water (W/O/W) emulsion consists of internal water droplets dispersed in larger oil droplets, which are themselves dispersed in an external continuous aqueous phase. On the other hand, an oil-water-in-oil (O/W/O) emulsion consists of oil droplets dispersed in water droplets, which are themselves dispersed in a continuous oil phase [4]. Nano-emulsion based food delivery systems (FDS) and drug delivery systems (DDS) have been applied in food, pharmaceutical and cosmetic fields. In the human digestive system, ingested food is broken down into small nutrient molecules that can be absorbed into the blood plasma. During the digestion process, most of the nutrients are transported as O/W emulsions. In particular, the particle size can be decreased from nano to mixed micelles order in the just before absorp-

(Received 5 Aug. 2015; accepted 17 Nov. 2015)

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*This article is based on a study first reported in the FFI journal Vol. 219, No. 4 (2014) with full reference.

tion. It is considered that high ratio of functional food compounds recovered from mixed micelles may have a high availability. However many of these compounds are unstable to heat, light, and oxygen; also, most of them are water-insoluble. The low solubility of these functional food compounds impairs their bioavailability and limits their use. On the other hand, the formulation of functional food compounds into micro/nano-emulsions is expected to improve their bioavailability. In this article, we present recent trends on the encapsulation of functional food compounds, focusing on the preparation and characterization of micro/nano-emulsion containing functional food components. We also present some applications of micro/nano-emulsions in food and other related industries.

2. Encapsulation of functional compounds

In general, there are two methods to produce micro/nano-emulsions with smaller droplet diameters and narrow size distributions: low- and high-energy input emulsifications. In high-energy emulsification methods, the emulsification is done using mechanical equipment including high-speed mixer, microfluidization, and ultrasonic emulsification. On the other hand, in low-energy emulsification methods, such as membrane emulsification, micro/nano channel emulsification, phase inversion emulsification and so on, the emulsions can be produced based on physicochemical behavior of the emulsifiers. We describe the process for using each approach in detail below. Depending on the methods employed, many types of emulsion systems containing functional com-

pounds were prepared, and investigated. Table 1 shows the summary of micro/nano-emulsions containing functional food compounds research and applications [5–17]. Besides, oil-in-water emulsion system using virgin *Picholine* olive oil, containing high flavonoids such as apigenin, showed higher antiallergenic effect compared with bulk olive oil system [18, 19].

2.1 High-speed mixer

High-speed mixers are used worldwide as emulsification devices in laboratory and industry fields. Figure 1 schematically represents details of a commercial high-speed mixer with rotor-stator polytron. The rotor-stator shaft consists of a rotor housed concentrically inside the stator with blades and a stator with different type of slots (vertical or slant). As the rotor rotates, it generates a

Fig. 1 High-speed mixer (Polytron®).

Table 1 Summary of research and application for micro/nano-emulsion loaded functional compounds.

Functional Compounds	Benefits	Applications	Reference
α -Tocopherol	Antioxidant	Medicine/Food	5
β -Carotene	Antioxidants/Vitamin A precursor	Food color	6
Curcumin	Antioxidant/Anti-inflammatory/Anticancer	Food color	7
Phytosterol	Cholesterol absorption inhibitor	Medicine/Cosmetic/Food additive	8
Lycopene	Antioxidant/Anticancer	Food color	9
Lidocaine	Local anesthetic/Antiarrhythmic drug	Medicine	10
Itraconazole	Antifungal	Medicine	11
Carbamazepine	Anticonvulsant	Medicine	12
Quercetin	Antioxidants/Anti-inflammatory	Medicine/Food	13
Cisplatin	Anticancer	Medicine	14
Thalidomide	Immunomodulatory agent	Medicine	15
Polyunsaturated fatty acids (PUFA)	Anti-inflammatory	Food/Medicine	16
Oleuropein	Antioxidant	Food	17

lower pressure to take the liquid in and out of the assembly, thereby resulting in cavitation and emulsification. High-speed mixer has processing capacity with broad ranges, typically from 0.005 to 10 L/h can be used for laboratory scale, and 10 to 5000 L/h are often used for industrial scale. The droplet diameters can be decreased with longer emulsification times and higher rotation speeds. However, the higher rotation speeds can also generate heat, therefore the temperature control is needed for some functional compounds that are sensitive to temperature [1]. Jafari *et al.* prepared emulsions containing essential oil by high-speed mixer. The mean droplet diameters were $10 \pm 0.08 \mu\text{m}$ [20].

2.2 High-pressure emulsification

A large amount of mechanical energy needed for the formation of micro/nano-emulsion. The free energy required (ΔG) for the area of oil–water interface increase (ΔA) can be given by the following equation.

$$\Delta G = \gamma \Delta A \quad (1)$$

where γ is the interfacial tension. For example, to prepare an O/W emulsion as γ is 10 mN/m when the volume fraction of oil is 0.1 and the radius is $0.1 \mu\text{m}$, the increased surface energy is calculated as 30 kJ/m^3 . Actually, small size of droplets can be formulated by oil droplet disintegration that requires a large pressure gradient, usually 1000 times of the normal value (30 MJ/m^3), to prepare such an emulsion. Moreover, the flow of a liquid through high shear rates results in large amount of energy dissipation, due to the friction losses. The excess energy is converted into heat, this is why cooling is required during emulsification. The effects of number of passes on emulsion temperature for different pressure levels are shown in Fig. 2. Seemingly, Fig. 3 shows the effect of emulsifying pressure on emulsion temperature at the exit of the valve. Temperature rises as the number of pass increases at the same pressure. Moreover, the temperature increases to around 80°C when the pressure is 300 MPa [21, 22].

High-pressure homogenization is the widely used method for preparing micro/nano-emulsions. High-pressure homogenization is extensively used both in laboratories and as well as in industries. Typical, often utilized high-pressure homogenizers are high-pressure valve homogenizer (HPH) and Microfluidizer. A HPH consists of a piston pump and a narrow gap, where the maximum operating pressure is 350 MPa. Droplet breakup occurs within the region of valve gap and in the

Fig. 2 Effects of number of passes on the temperature of emulsions, for different operating pressure [21].

Fig. 3 Effect of pressure on temperature at the exit of the valve [22].

jet after the gap (Fig. 4) [23]. Figure 5 shows that an emulsion containing β -carotene was produced by HPH. Here, β -carotene was dissolved in medium chain triacylglycerol as a dispersed phase and 10% of Tween 20 aqueous solution was used as continuous phase. The processing was carried out 3 times for each pressure, and the droplet diameter tended to decrease with the pressure increase [24]. Besides, some factors can lead to the decrease of the droplet diameter such as the increased

Fig. 4 High-pressure valve homogenizer [23].

Fig. 5 Preparation of β -carotene loaded nanoemulsions by high-pressure emulsification [24].

Fig. 6 Schematic representation of Microfluidizer[®] interaction chamber.

number of passes, the decrease of interfacial tension and if the viscosity ratio of disperse phase to continuous phase is between 0.05 and 5 [25]. Table 2 summarizes some references where HPH was used [24, 26–28]. On the other hand, Microfluidizer[®] comprises of a fluid inlet, a pump, and a chamber with two channels where the fluids are taken into, and then impact with each other (Fig.6). The coarse emulsion is forced into an inlet channel by high pressure and divided into two streams to make an impact force on each other at high velocity in the interaction chamber. The intensive shear force generated in the interaction chamber is highly effective for breaking droplets up and forming 30 nm – 3 μ m-order micro/nano-emulsions. Table 3 summarizes references related to the emulsion preparation using Microfluidizer [29–35]. The droplet diameter can also be affected by increasing the operating pressure, the number of passes and so on.

2.3 Ultrasonic emulsification

Ultrasonic emulsification is also a very effective processing method in the generation and application of

Table 2 An overview of reference on O/W nano-emulsion formulation using high-pressure valve homogenizer.

Dispersed Phase**	Functional Compounds	Emulsifier*	Droplet Diameter (nm)	Reference
MCT	β -Carotene	Tween 20, 40, 60, 80	114–178	24
MCT	β -Carotene	Tween 20, ML750, WPI, Modified starch	115–303	26
MCT	Curcumin	Tween 20	82–184	27
Olive oil	–	Pluronic [®] F-68, Phospholipid	239–274	28

*Tween: Polysorbate

*ML: Decaglycerol monolaurate

*WPI: Whey protein isolate

**MCT: Medium chain triglyceride

Table 3 An overview of references on O/W nano-emulsion formulation using Microfluidizer[®].

Dispersed Phase**	Functional Compounds	Emulsifier*	Droplet Diameter (nm)	Reference
<i>d</i> -Limonene/Fish oil	–	WPC, WPH, SC, Modified starch	175–585	29
Peanut oil	–	Tween 80	115–303	30
MCT**	Coenzyme Q10	Non-ionic Surfactant, Lecithin, Tween 20, β -Lactoglobulin	60	31
MCT**	α -Tocopherol	Lecithin	85–381	32
Corn oil	–	Tween 20, β -Lactoglobulin	60–150	33
Soybean oil	–	Polyglycerol esters of fatty acids	180–380	34
Soybean oil	β -Carotene	Polyglycerol esters of fatty acids	170–270	35

*Tween: Polysorbate

*WPC: Whey protein concentrate

*WPH: Whey protein hydrolysate

*SC: Sodium caseinate

**Medium chain triglyceride

emulsions. In ultrasound emulsification, the energy input is provided by sonicator probe. The sonicator probe is placed in the target liquids, where the intense mechanical vibrations are generated, leading to cavitation effects. The collapse of the micro bubbles occurs by cavitation forces in the immediate vicinity of the sonicator probe that leads to droplet disruption. Ultrasound can be generated by mechanically and electrically. A schematic diagram of an ultrasonic emulsification is shown in Fig. 7. In general, 1–30 μm order of emulsions can be produced by ultrasonic emulsification. Recently, it has been reported that nano-order of emulsions can also be produced by ultrasonic emulsification (Table 4) [36–39]. The droplet diameter of produced emulsion depends on the ultrasonic parameters such as irradiation intensity, irradiation time, emulsifier type and concentration, among others [36].

2.4 Membrane emulsification

Membrane emulsification (ME) was proposed by Nakashima *et al.* [40]. In membrane emulsification process, the microporous membranes were used for produce droplets by pressing the dispersed phase through

Fig. 7 Schematic representation of ultrasonic homogenizer.

the pores of microporous membrane directly into the continuous phase (Fig. 8) [40]. Compared to conventional emulsification, the advantages of ME can be carried out with lower energy input to generate smaller sized droplets and narrow droplet size distributions. The droplet diameter depended on the pore size of porous membranes, and the coefficient of variation on size of the droplets produced was about 10%. The different types of ME system and emulsified case are shown in Table 5 [41–46]. Membrane emulsification is applicable to O/W emulsions, W/O emulsions and multiple emulsions. Shirasu porous glass (SPG) is a commonly used membrane for emulsion preparation. In general, the droplet diameters were determined to be 3–10 times bigger than the mean diameter of the membranes pores. On the other hand, premix emulsification can also be used to produce emulsions with high dispersed phase fraction. In this case, the droplet diameters may be 1–2 times larger than the mean diameter of the membranes pores.

2.5 Emulsification via micro/nano engineered devices

Microfluidic devices available for flowing fluids on a

Fig. 8 Schematic representation of membrane emulsification using Shirasu porous glass (SPG) membrane.

Table 4 An overview of reference on O/W nano-emulsion formulation using ultrasonic homogenizer.

Dispersed Phase	Emulsifier*	Droplet Diameter (nm)	Reference
Sunflower oil/ Canola oil	Tween 80	40	36
Linseed oil	Tween 40	150	37
Olive oil/ Sesame oil/ Soybean oil	Pluronic® F-68	<500	38
Soybean oil	Phosphatidylcholine, Sodium palmitate, Sucrose palmitate	58	39

*Tween: Polysorbate

Table 5 Different type of membrane emulsification systems and applications.

Membrane type and material	Process Design	Dispersed Phase	Emulsifier*	Droplet Diameter (μm)	Reference
Tube membrane (SPG, Ceramic)	Cross flow	Sunflower oil	Tween 20, 80, Lecithin, WPI	2.6–6.5	41
Tube membrane (Micro-porous glass)	Stirred cell	Sunflower oil	Tween 20	40–400	42
Flat membrane (Micro-porous glass)	Stirred cell	Pumpkinseed oil / Sunflower oil	Tween20, Pluronic® F-68	74–273	43
Flat membrane (Stainless steel)	Rotating flat	Paraffin	Tween 20	79–259	44
Tube membrane (Stainless steel)	Vibrating and rotating	Sunflower oil	Tween 20	30–50	45
Flat membrane (SPG, Polyether sulfone)	Premix	Corn oil	Tween 20, 80	1–12	46

*Tween: Polysorbate

*WPI: Whey protein isolate

chip is an emerging and multidiscipline technology. It was developed in the early 1990's. Emulsification techniques via microchannel (MC) devices have been proposed for producing mono-disperse emulsions in the last two decades [47]. Emulsification via micro devices is summarized in Table 6 [48–53, 55]. We presented below a brief introduction about each devices:

T- and Y-junctions are frequently used microfluidic geometries to produce mono-dispersed droplets by using the cross-flow shear method. The cross-flow shear method utilizes shear force, imposed by an immiscible cross-flow fluid to generate droplets (Fig. 9a and 9b) [48, 49]. The flow-focusing geometry implemented in a microfluidic device is shown in Fig. 9c. A liquid flows into the central channel and a second immiscible liquid flows into the two outer channels. The two liquid phases are then forced to flow through a small orifice that is located downstream of the three channels. The outer fluid exerts pressure and viscous stresses that force the inner fluid into a narrow thread, which then breaks inside or downstream of the orifice. Figure 9d shows a glass capillary device consisting of two round glass capillaries lying end-to-end within a square glass tube. When the two fluids enter the collection tube, emulsion droplets are generated without chemical surface modification,

in which the droplets obtained are smaller than the size of the collection tube. Multiple emulsion can also be prepared by this device [51].

Microchannel (MC) emulsification is also a technique for producing monodispersed emulsions in which droplets are formed by spontaneous transformation caused by interfacial tension. It can be divided into grooved-type MC plate and straight-through [52–53]. Grooved-type MC plate is shown in Fig. 9e, parallel grooves fabricated on a terrace are arranged on four sides of the plate. The dispersed phase supplied through a central hole is forced to flow through MCs, because the plate is sealed with a transparent cover plate. The dispersed phase takes a disk-like shape on terrace and this shape is characterized by a higher interfacial area per unit volume than a spherical shape, resulting in hydrodynamic instability. This instability is a driving force for spontaneous transformation of dispersed phase into spherical droplets. Asymmetric straight-through MCs is shown in Fig. 9f. It has circular channels on the upstream (bottom) side and slots on the downstream (top) side. Asymmetric plate is useful for low viscosity oils (less than 1 mPa s). The mean droplet diameter of emulsion produced by asymmetric plate can be controlled by shape and size of MC. A 30 μm -order of polyunsaturated fatty acids (PUFA)-

Table 6 Summary of emulsification via micro devices.

	Device type	Monodispersity	Droplet Diameter (μm)	Productivity (mL/h)	Fabrication	Reference
Micro fluidics emulsification	T / Y junction	Good	50–500	1–300	Simple	48, 49
	Flow focusing	Good	–100	1–50	Elaborate	50
	Micro capillary	Good	–10	1–10	Elaborate	51
Microchannel emulsification	Grooved microchannel	Excellent	1–500	1–10	Simple	52
	Asymmetric straight through microchannel	Good	5–2000	1–1000	Elaborate	53
	Edge-based droplet generation	Good	5–10	1–10	Simple	54

(a) T junction, (b) Y junction, (c) flow focusing, (d) micro capillary, (e) groove microchannel, (f) asymmetric straight through microchannel, (g) edge-based droplet generation

Fig. 9 Different type of micro devices which can produce monodispersed droplets [48–53, 57].

loaded O/W emulsions and high concentration of vitamin C-loaded multiple emulsions have been produced by asymmetric straight-through MCs [54, 55]. Recently nano-channel has also been developed, and monodispersed emulsions with the mean droplet diameter of approximately 500 nm have been produced [56].

Edge-based droplet generation (EDGE) emulsification was proposed by van Dijke *et al.* [57], where simultaneous formation of many oil droplets in a single EDGE module occurs. In EDGE module, multiple droplets are formed at the edge of a shallow but rather wide rectangular plateau (Fig. 9g).

2.6 Phase inversion emulsification

Some methods have been developed to produce nano-emulsions by inducing a phase inversion in emulsions from a W/O emulsion to O/W emulsion or vice versa, such as phase inversion temperature (PIT), phase inversion composition (PIC), and emulsion inversion point (EIP) methods [58–60]. All these methods depend on the changing of the spontaneous curvature of the surfactant. For non-ionic surfactants, the hydrophilic-lipophilic bal-

ance (HLB) can be changed with temperature. An O/W emulsion formed at low temperature, and a W/O emulsion at higher temperature. During cooling, the system crosses a point of zero spontaneous curvature and minimal surface tension, promoting the formation of finely dispersed oil droplets. This method is called PIT method (Fig. 10). It was reported that an emulsion with droplet diameter around 100 nm can be produced using this method [61]. Here, the stability of resulting emulsions was greater and depended on the storage temperature. Therefore, the rapid cooling is required after producing the emulsion [62]. On the other hand, a PIC method starts from W/O emulsion, which is an equilibrium phase of oil, water, and surfactant, in which the surfactant is already at the water/oil interfaces with a curvature turned toward water. Then a large addition of water, which causes an inversion of the spontaneous curvature of the emulsifier, now turned toward the oil. After that, an emulsion with diameters of around 100 nm can be obtained (Fig. 11). Both PIT and PIC methods are reversible. In contrast, in case of EIP method, the change from W/O emulsion to O/W emulsion or vice-versa is

Fig. 11 Schematic representation of phase inversion composition (PIC).

Fig. 10 Schematic representation of phase inversion temperature (PIT).

through a catastrophic phase inversion. In this case, a W/O emulsion with a high oil to water ratio is formed using a low molecular weight emulsifier, and then increasing amounts of water are added to the system with continuous stirring (Fig. 12). Above a critical water content, the water droplet concentration is higher than the emulsion reaches a phase inversion point where it will change from W/O emulsion to O/W emulsion. The droplet diameter produced by EIP method depends on process variables (stirring speed, rate of water addition). It has been reported that an emulsion with droplet diameter around 100 nm can be produced by EIP method [63].

Fig. 12 Schematic representation of emulsion inversion point (EIP).

2.7 Solvent displacement/emulsification evaporation

Figure 13 shows the schematic representation of solvent displacement method. Initially the functional compounds are dissolved in an organic solvent e.g., alcohol or acetone as an oil phase, while the oil phase is added to the aqueous phase drop-by-drop under magnetic stirring leads to a dispersion. After this step, some water is added to the dispersions, in order to allow the diffusion of the organic solvent into the water. Additional evaporation process is used to remove the organic solvent from the dispersions. Emulsification evaporation is also one of the most common methods used for preparing nano-dispersions. With this method, the functional compounds

Fig. 13 Schematic representation of solvent displacement method.

are dissolved in a lipophilic solvent e.g., hexane as an oil phase, and an O/W emulsion is formed by conventional emulsification with the aqueous phase containing an emulsifier. Converting the emulsion into nano-dispersion is then carried out by evaporating the solvent, and the particle size decreases to the nano-size range (Fig. 14). Table 7 summarizes some references where solvent displacement/emulsification evaporation methods were used [64–67]. The particles produced using these methods may have from several 10 nm to 200 nm-order.

2.8 Spontaneous emulsification

Spontaneous emulsification is also one of methods which can prepare nano-sized emulsion. An emulsion is spontaneously formed when two liquids such as organic phase and aqueous phase are mixed together at a special temperature. Spontaneous emulsification was depended on the chemical composition of phases and the presence of surfactants. Figure 15 shows the schematic represen-

tation of spontaneous emulsification [68–69]. Initially, the organic and aqueous phases are mixed together. After mixing, the water-miscible solvent and surfactant move from the organic phase into the aqueous phase, which creates a large turbulent force at the oil water interface. Some references using spontaneous emulsification are summarized in Table 8 [70–72].

Fig. 14 Schematic representation of solvent evaporation method.

Fig. 15 Schematic representation of spontaneous emulsification.

Table 7 An overview of literature researches on oil in water nano-emulsion formation using solvent displacement and evaporation methods.

Dispersed Phase	Functional Compounds	Emulsifier*	Droplet Diameter (nm)	Reference
Hexane	β -Carotene	Polyglycerol esters of fatty acids	60–140	64
Hexane	β -Carotene	Casein sodium	18	65
Corn oil/Ethyl acetate	–	Whey protein	<70	66
Acetone	β -Carotene	Tween 20	77	67

*Tween: Polysorbate

Table 8 An overview of literature researches on oil in water nano-emulsion formation using spontaneous emulsification method.

Dispersed Phase	Functional Compounds	Emulsifier*	Droplet Diameter (nm)	Reference
MCT**	α -Tocopherol	Span 80, 85, Tween 20, 80	100–600	70
MCT**	Oligothymidylate	Lecithin	225	71
Palm oil	Vitamin E	Pluronic® F-68	94	72

*Span: Sorbitan monooleate

*Tween: Polysorbate

**Medium chain triglyceride

3. Characterization of micro/nano-emulsions

Emulsion are thermodynamically unstable systems that tend to breakdown over time through different physicochemical mechanisms [1]. Here, we will mainly focus on the physical stability (Fig. 16). The physical stability of emulsion can be evaluated by Stokes' law equation:

$$V_{\text{stokes}} = \frac{2r^2(\rho_0 - \rho)g}{9\eta_0} \quad (2)$$

where, V_{stokes} is creaming velocity, r is the droplet radius, ρ is droplet density, ρ_0 is the density of continuous phase, η_0 is the viscosity of continuous phase, and g is the gravitational acceleration. In here, the V_{stokes} value is negative for creaming, and positive for settling. This equation shows that the creaming can be affected by droplet radius, viscosity of continuous phase and density difference between the oil and water phases. On the other hand, the stability of an emulsion can also be evaluated by measuring the change in droplet diameter when the emulsion is stored for a specified length of time, or exposed to some environmental stresses, such as heating, freezing and stirring. The parameter named emulsion stability index (ESI) based on droplet diameter measurements, is given below:

$$\text{ESI} = \frac{d_0 \times t}{d_t - d_0} \quad (3)$$

where, d_0 is the initial mean droplet diameter of the emulsion and d_t is the mean droplet diameter measured at time t [1]. It should be mentioned that there are many

potential problems associated with any methods used to define the ESI for comparing the effectiveness of different types of emulsifiers. Increases on droplet diameter in an emulsion may be caused by different mechanisms, such as flocculation, coalescence, or Ostwald ripening. It is important to establish which mechanism is dominant in a particular system. The mean droplet diameter does not usually increase linearly with the time, so that the value of ESI may depend on the time during the measurements. It is therefore important to use a standardized storage time to compare the emulsifier effectiveness, for instance, 1 day or 1 week. The particle size distribution may also shift from a single peak to multi-peaks with the storage time. In some cases, the changes in particle size distribution with time are more important than the mean droplet diameter. In addition, the ESI strongly depends on the type of mean droplet diameter. At present, there is no single parameter, such as ESI, which can be used to quantitatively compare the effectiveness of emulsifiers. Therefore, the effectiveness of emulsifier on stabilizing droplets against aggregation is usually evaluated by the mean droplet diameters or droplet size distributions, rather than by a calculated ESI. However, the ESI is more effective compared to a series of emulsifiers under the same conditions.

For micro/nano-emulsion containing functional compounds, both the emulsion stability and functional compounds stability have to be evaluated. Spectrophotometer and HPLC are the main equipment used to evaluate these parameters. Moreover, the bioavailability and toxicity of emulsion containing functional compounds also have to be evaluated by *in vitro* gastrointestinal model, animal testing or in some cases, by human test.

4. Application of micro/nano-emulsions in food

Micro/nano-emulsions can be used in many fields. There is increasing interest in food industry in utilizing them as delivery carriers containing lipophilic functional food compounds (*e.g.*, vitamin A, D₃, E, polyphenols, carotenoids, phytosterols, and flavor compounds) to protect and deliver those compounds, besides from increasing the bioavailability of those lipophilic compounds. For instance, citral is one of the most important flavor compound that is widely used in the food and beverage industries. Citral is chemically unstable and degrades over time in an acidic environment; also can be affected by heat, light and oxygen [73]. It has been reported that the stability of citral can be improved by emulsification

Fig. 16 Schematic representation of emulsion destabilization.

Table 9 Commercial nano-emulsion formulations.

Commercial Name	Manufacturer	Reference
Canola Active Oil	Shemen Industries, Israel	http://www.nanotechproject.org/cpi/products/canola-active-oil/ (20140908)
NanoResveratrol TM	Life Enhancement, USA	http://jp.iherb.com/life-enhancement-nanoresveratrol-1-7-fl-oz-50-ml/8975 (20140908)
Nutri-Nano TM CoQ-10 3.1x Softgels	Solgar, USA	http://simplysolgar.co.uk/collections/coenzyme-q-10 (20140908)
Spray for Life [®] Vitamin Supplements	Health Plus International [®] , Inc., USA	http://www.nanotechproject.org/cpi/products/spray-for-life-r-vitamin-supplements/ (20140908)
Spray for Life [®] Vitamin Supplements	Health Plus International [®] , Inc., USA	http://www.nanotechproject.org/cpi/products/spray-for-life-r-vitamin-supplements/ (20140908)
Omega 3 Bread	Tip-Top Bakery, Australia	http://www.tiptop.com.au/healthnutrition/nutrition-fact-sheets/omega-3 (20140908)
Color particles	Fujifilm Co., Japan	http://astalift-white.jp/technology_nano.html (20140908)

method [74, 75]. Phytosterol can reduce cholesterol levels in humans. Due to the water insolubility, incorporation of phytosterol into foods is difficult. Recently, some research groups produced emulsions containing high concentrations of phytosterol [76–78]. In addition to this, the encapsulation of lycopene [79], astaxanthin [80], coenzyme Q10 [81] also has been reported by different research groups. Some reports using nano-emulsions are also summarized in Table 9.

5. Conclusions

This article reviews on the current status of micro/nano-emulsion containing functional food components preparation and characterization, and presents some applications of micro/nano-emulsions into food industry.

Recently, different types of devices and methods have been developed to produce micro/nano-emulsions. We mentioned some of them in this paper. Micro/nano-emulsion based materials have a large surface area-to-volume ratio. In the future further applications, such as the development of new emulsification systems, as well as functional foods delivery systems or drug delivery systems, are expected.

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◆◆◆ 和文要約 ◆◆◆

食品機能性成分含有マイクロ／ ナノエマルジョンの作製と特性評価

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エマルジョンとは水-油のように、互いに混じり合わない2種の液体で、一方が他の液体中に分散しているもののことをいう。互いに混じらない水-油を乳化剤の作用で細かい液滴として分散させる操作を乳化といい、乳化する作用をもつ物質を乳化剤という。同一組成のエマルジョンにおいては、分散液滴径と液滴径分布がエマルジョンの諸特性に大きく影響を与える。分散液滴の液滴径が小さく、液滴径分布が狭くなるほどエマルジョンの安定性が向上する。エマルジョンの粒子径は通常 0.01 ~ 100 μm 程度であり、20 ~ 200 nm 程度の微細な粒子径のエマルジョンはナノエマルジョンとよばれている。水-油系エマルジョンを形成する場合、油滴が水に分散する水中油滴 (O/W) 型エマルジョンと、水が油に分散する油中水滴 (W/O) 型エマルジョンなどがあげられる。また、水中に分散している油滴がさらに水を内包しているような状態は Water-in-Oil-in-Water (W/O/W) 型エマルジョンといい、これに対して、油中に分散している水滴がさらに油相に分散させた状態を Oil-in-Water-in-Oil (O/W/O) 型エマルジョンという。ナノエマルジョンは食品機能性成分送達システム (FDS) やドラッグデリバリーシステム (DDS) として食品、医薬品、化粧品分野への

応用が検討されている。普段摂取した食品は、消化管で様々な消化液と混合し細かく分解され、体内に吸収された後に主として生体膜における生体反応に関与するが、摂食から吸収までの消化過程においては、O/W エマルジョン粒子として運搬されている。とくに、吸収直前では、粒子サイズがナノメートル程度のナノエマルジョンから混合ミセルまで粒子サイズが微細化すると考えられている。混合ミセルへの機能性を有する成分の取込率が高いことを体内吸収性が高いとみなせる。ポリフェノール類やカロテノイド類を含む食品機能性成分は、健康の維持や病気の予防に有効であることが知られているが、その多くは熱、光、酸素などに対して不安定であり、また脂溶性のものが多いため、生体利用性が低いのが現状である。エマルジョン化することで、脂溶性の食品機能性成分の生体内への吸収性が改善できると知られているが、ここで、近年、食品機能性成分を含有するマイクロ／ナノエマルジョンの作製法の事例およびその特性評価について紹介する。

なお本稿は、Foods & Food Ingredients Journal of Japan (FFI ジャーナル) 219 巻 4 号 312 - 323 頁 (2014 年) に掲載された原稿を、許可を得て、英訳したものである。

(受付 2015 年 8 月 5 日, 受理 2015 年 11 月 17 日)

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