A review on natural-based active compounds delivery system and its potential in food preservative application

N A F M Anuar, F Kormin, and N A Z Abidin

Department of Technology and Heritage, Faculty of Applied Science and Technology, University Tun Hussein Onn Malaysia, Edu Hub Pagoh, Km1, Jalan Panchor, 84600 Pagoh Muar Johor.

Abstract. Most of the essential oils extracted from various plants has a great potential to be used as a natural source of antimicrobial agent. These natural antimicrobials compounds are poorly soluble or lipophilic and thus required surfactant and co-surfactant to make them stable and boost up their efficacy as a food preservative. Regarding to these problems colloidal delivery systems based on microemulsions or nanoeemulsions are increasingly being utilized in the food and pharmaceutical industries to encapsulate, protect, and deliver lipophilic bioactive components affectively. Unfortunately, in major foods and pharmaceutical industry, ternary food grade surfactant (such as tween-20, tween-80 span-80) were commonly being selected to be used along with pH adjustments compared to another available natural surfactants such as sodium caseinate, gelatin and lecithin. The objective of this review is to present briefly the possible applications of these novel systems of nanoeumulsion and microemulsions using recent methods and techniques. Review on journal related on formation of emulsion systems applied in industries especially on foods and beverages had been done. For example, review had been done on journal discussed on application of essential oil as a flavor in beverages, application of microemulsion system as natural food coloring in food products as well as oil loaded microemulsion and its potential to be used as a natural food preservative. All method used in the journal has been reviewed, analyzed and improvised for a better further application.

1. Introduction

Processed, packaged, refined, and conveniences foods were directly related with food sustainability issues. These strategies had been used to prolong shelf-life, freshness, improve and maintained nutritional values of food product, assist in transportation as well as to overcoming food starving. Because of the costing, stability and availability issues, artificial and synthetic preservatives being highly demanded by most of the food manufactures compared to application of natural preservatives. Nowadays, hundreds of approved additives had been listed by the U. S. Food and Drug Administration (FDA). Unfortunately, the studies on its safety for consumption that existed were often inadequate to assess the degree of health risks. Food toxicity and its health-related issues are becoming nearly every day occurrences, leaving consumer to wonder if our food system is designed to kill us.

However, the main constrains in using natural food preservatives were its stability, availability and production cost. As for example, antimicrobial properties of lemongrass essential oil increase their potential to be use as natural preservatives for food products. Unfortunately, it will be unstable (separation and cloudiness occur) once simply added into beverages and another water-based product. These stability problems influenced the effectiveness of antimicrobial activities towards product system. Furthermore, oil extraction process involved high technology machines with high electrical voltages and
bulk raw materials required for small amount of output. These processes increased the value and market price rate. Regarding to these issues, application of emulsion helps in delivering high-value functional ingredients in the food system efficiently. Through encapsulation and dispersion technologies, the optimal targeted performance at the lowest concentrations would be achieved. Thus, study on development and application of stable food preservative for specific food product involving various sources of natural antimicrobials were in increasing-trend.

2. Types and characteristics of emulsion applied in food products

Emulsions technology known as microemulsion, nanoemulsion, and microemulsion could be developed into either water in oil (W/O) emulsion or oil in water (O/W) emulsion types (Refer figure 2). Figure 1 showed the specific size range for each emulsion groups. Each droplet sizes and types of emulsion will directly affect their suitability in food application. As for example, water in oil emulsion suited for oil-based food such as butter cream while oil in water emulsion suited for water-based product such as beverages.

![Figure 1. Types and typical droplet diameter sizes of macroemulsion, nanoemulsion and microemulsion. Sources: [1].](image1)

![Figure 2. Schematic representation of water in oil (left) and oil in water (right) macroemulsions. Sources: [1].](image2)

2.1. Macroemulsion

Macroemulsion was characterized under the largest size range of emulsion droplet. Droplet size of macroemulsion is larger than 400nm which make it grouped under course emulsion. Physically, it was characterized with creamy, sticky but flowable liquid. It can be form into either water in oil (W/O) or oil in water (O/W) types.

2.2. Nanoemulsion

Nanoemulsion was characterized under the medium size range of emulsion droplet. Droplet size of macroemulsion was ranged from 100nm to 400nm. Physically, it was characterized with either translucent or transparent and flowable liquid. Compared to microemulsion, nanoemulsion is thermodynamically unstable as they would react with surrounding temperature exchanged hence affect their cloudiness. For example, nanoemulsion might be affected by heat exchanges during heat treatment in food preparation process. During heating and high-shear rate mixing, nanoemulsion would be appeared slightly turbid in color hence become transparent once fully cold-down. It was completely different with microemulsion system that able to stay transparent and static with 1 phase of liquid in various surrounding temperature.

2.3. Microemulsion

Microemulsion was characterized under the smallest size range of emulsion droplet. Droplet size of microemulsion was ranged from 10nm to 100nm. Physically, it was characterized as transparent and flowable liquid. It can be form into either water in oil (W/O) or oil in water (O/W) microemulsion types.
Microemulsion was thermodynamically stable thus make it physically stable under various temperature conditions. Besides, compared to another emulsions, microemulsion is very easy to prepare and scale up due to spontaneous formation ability. This system raised the absorption rate of bio-active compounds as well as boost-up bio-availability by eliminating interfering variations. Besides, this system improved solubility of lipophilic bio-active compounds. It thermodynamically stable characteristics make them more stable compared to conventional and hence suitable for long term use. Unfortunately, additional use of excess amount of surfactant and co-surfactant increases cost of preparation. Furthermore, excess concentration of surfactants can lead to mucosal toxicity.

3. Major components of emulsion

Perfect emulsion formation might include interaction between three major components which are oil, surfactant as well as co-surfactant. Table 1 below summarized the emulsion’s component and their examples.

| Oil | Surfactant | Co-surfactant |
|-----|------------|---------------|
| Saturated fatty acid-lauric acid, myristic acid, capric acid | Polyoxylethylene/Polsorbate/Tween 20,40,60,80 | Ethanol, propanol, Isopropanol, butanol, pentanol, hexanol, sorbitol, n-pentanoic acid, n-hexanoic acid, n-butyamine, sec, butyamine, 2-amino pentane, 1,2-butanediol, Propylene glycol. |
| Unsaturated fatty acid-oleic acid, linoleic acid, linolenic acid | Sorbitan Monolaurate (Span) | Cremophor RH40 (polyoxyl 40 hydrogenated castrol oil) |
| Fatty acid ester-ethyl or methyl esters of lauric, myristic and oleic acid. | Soybean lecithin | Pluroroleique (polyglyceryl-6-dioleate) |
| Example: (GlycerylMonooanddicaprate, isopropylmyristate, sunflower oil, soyabean oil, Labrafac CC), surfactant (Cremophor EL, Labrasol) | Lyso lecithin | Pluroniosstearique (isostearic acid of polyglycerol) |
| | Sodium dodecyl sulphate (SDS) | Distearoylphosphatidyl ethanolamine–N–poly (ethyleneglycol)2000 (DSPE–PEG) |
| | Sodium bis (2–ethylhexyl) sulphosuccinate (Aerosol OT) | Poloxamer |
| | Dioctyl sodium sulphosuccinate | Polyoxyethylene–10–oetyl ether (Brij 96V) |
| | Sodium deoxycholate | Polysorbate 80 (Tweens80) |
| | Labrasol (Polyethylene glycol–8–caprylic acid) | Span 20 |
| | TritonX–100 | Sodium monohexyl phosphate |
| | | Sodium monoocetyl phosphate |
| | | N,N–Dimethyl dodecylamine–N–oxide (DDNO) |
| | | N,N–Dimethyl octylamine–N–oxide (DONO) |
| | | Cinnamic alcohol |
| | | Cinnamic aldehyde |

Sources [2], [3], [4], [5], [6] & [7]

3.1. Oil
Oil is a lipophilic bio-active compound that completely blended in emulsion system, which then help them to be safely and effectively delivered into a targeted food system hence act as a food preservative. In emulsion technology for development of natural food preservative, organic and lipophilic essential oil widely being used as a source of natural antimicrobial agent in processed foods. Through emulsion technology, antimicrobial compound such as thymol and limonene from essential oil could be added into beverages product by improving the oil stability. Besides, trending in using essential oil as a food preservative was directly related to food safety and awareness. Oil loaded emulsions is being used as an alternative of synthetic food preservative as well as to improve food safety.

3.2. Surfactant
In general, surfactant is a substance that help to reduced interfacial tension between two immiscible liquids in colloidal system. Surfactant could be found from natural sources or artificially produced and it has been used to promote the physicochemical stability of major food system. Normally, industries used artificial surfactant known as tween-80, tween-20, and span-80 in production of all oil-based liquid flavouring, coloring, and other food additives. With these surfactant, microemulsion and nanoemulsion system would be developed with high-stability of its physicochemical characteristics as well as increased its potential to be used as an efficient preservative compared to solo-application of lipophilic compounds.

3.3. Co-surfactant
Co-surfactant used to support surfactant’s function and improve the stability of colloidal system. It would help in dispersion of two immiscible liquids as well as give protection for carried bio-active compounds from being easily degraded during food processing phases. Thus, availability of bio-active compound would be long-lasting in food system and then efficiently preserve the foods or beverages well. Bioactive compound such as essential oils, usually consisted of volatile compound hence make them easily to be evaporated into the exposed air. Thus, through microemulsion system, the oil (with natural antimicrobial compounds) can be fully dispersed and homogenized in water-based product which then help in preservation efficacy.

4. Application of emulsion technology as a delivery system for lipophilic and high-value components in food and beverages.
In order to optimize preservative action of bio-active components from extracted oil, researchers studied on various type of potential surfactants and co-surfactants used for specific food and beverages product. They also came out with the list of preparation methods and conditions applied to develop a physicochemically stable of macroemulsion, nanoemulsion and microemulsion. Table 2 below summarized the recent application of emulsion technology as a delivery system for lipophilic and high-value components in various food and beverages product.
Table 2. Application of emulsion technology as a delivery system for lipophilic and high-value components in various food and beverages product.

| Type of emulsions/ Literature Sources | Application/Dispersion materials | Preparation methods | Effect in physicochemical stability of bio-active compounds |
|--------------------------------------|----------------------------------|---------------------|----------------------------------------------------------|
| **Microemulsion** [8]                | Dispersion of oil-based flavouring substance (β-carotene) in formulated peppermint-microemulsion. Tween-20 was used as a surfactant, while sunflower lecithin used as a co-surfactant | i. Dissolve β-carotene in peppermint oil at a mass ratio of 1:30.  
ii. Add Tween-20, lecithin, and distilled water to a specific total mass.  
iii. Prepare the coarse emulsions by hand-shaking the mixture for little seconds then heat it for 5 minutes, at 80 °C.  
iv. Hand-shaking the sample in ice/water bath to get Clear samples.  
v. Analyze Isotropic structure of microemulsions through the polarised light microscopy (Model BX51; Olympus Corp. of the Americas, Melville, NY). | A peppermint oil concentration of 3% was chosen because it was the highest amount of oil that was dissolved by 20% Tween-20.  
Particle size with <10 nm obtained.  
Microemulsion was stable under room temperature storage for 65 days produced.  
Less degradation of β-carotene in microemulsion during various beverages process conditions. |
| **Microemulsion** [9]                | Dispersion of various percentage of β-carotene microemulsions using Span 80, Span 40, Tween 80 as ternary food grade surfactant with either virgin coconut oil or palm oil as oil phase. Microemulsion development through spontaneous emulsification method.  
i. Mix all ternary food grade nonionic surfactants mixture.  
ii. Use phosphate buffer as aqueous phase.  
iii. Add β-carotene in mixed surfactant-oil, heat and stir using heating magnetic stirrer (AREC, VELP Scientifica, Italy) at 70ºC and 800 rpm. Wait for 10 minutes.  
iv. Add the aqueous phase dropwise while stirring and heating up to 20 minutes.  
v. The microemulsions were maintained at ambient temperature for 24 h to reach equilibrium before further investigation. | Microemulsion using palm oil as oil phase showed slower degradation of β-carotene during storage hence make it suitable to be utilize as delivery systems to encapsulate and stabilize β-carotene for food products.  
Particle sizes range from 10 – 50 nm obtained. [3] |
| Type of emulsions/ 
| Literature Sources | Application/Dispersion materials | Preparation methods | Effect in physicochemical stability of bio-active compounds |
|---------------------|----------------------------------|---------------------|----------------------------------------------------------|
| Microemulsion       | Study on potential of lecithin,  | i. Select monophasic | Highest physicochemical stability and antioxidant activity (0.93 mM trolox equivalents) obtained from gallic acid-loaded microemulsion as compared to other antioxidant carriers studied. |
| [10]                | caprylic/capric triglycerides,  | area of microemulsion |                                                          |
|                     | isopropyl myristate, alcohols and | through pseudo-ternary phase diagram |                                                          |
|                     | water for natural antioxidant (gallic acid, p- | determined at 25°C. |                                                          |
|                     | hydroxy benzoic acid, protocatechuic acid and | ii. Mix surfactants and co-surfactants together. |                                                          |
|                     | tyrosol) carrier.                   | iii. Obtain desired concentration of the antioxidant in |                                                          |
|                     |                                  | the microemulsions. Shake gently for less than 1 min.  |                                                          |
|                     |                                  | iv. Keep the obtained isotropic micellar solution at |                                                          |
|                     |                                  | 25°C. |                                                          |
| Microemulsion       | Study on potential of lecithin,  | i. Disperse phosvit in pH 7.0 buffer (10 mM PBS) | • The particle diameters of microemulsions containing 0.5 to 2.0% phosvitin were ranged from 0.4-2.7 m. |
| [11]                | caprylic/capric triglycerides,  | at room temperature to form phosvitin solutions. | • The emulsifying activity index increased when phosvitin concentration increased. |
|                     | isopropyl myristate, alcohols and | ii. Primarily dissolve resveratrol crystals with | • This study suggested to use phosvitin (egg protein) and polyphenol together prior to efficiently inhibit oxidation in food emulsions. |
|                     | water for natural antioxidant (gallic acid, p- | ethanol then mix the resveratrol solution with |                                                          |
|                     | hydroxy benzoic acid, protocatechuic acid and | peanut oil. |                                                          |
|                     | tyrosol) carrier.                   | iii. Prepare emulsion by mixing oil phase and phosvitin solution. Run the sample in high-speed homogenization at 10,000 rpm for 1 minute using a high speed blender (IKA T25 Basic, Staufen, Germany). |                                                          |
|                     |                                  | iv. To produce microemulsion, double the |                                                          |
|                     |                                  | homogenization process with a high-pressure homogenizer at 100 MPa through the ATS homogenizer (ATS Engineer Inc., Shanghai, China). Seal and store samples at 4 °C. |                                                          |
| Microemulsion       | Dispersions of lemon oil into | i. Microemulsion developed by titration methods. | Lemon oil composition will directly affect the colloidal formation system that have a potential to be used in food and beverages applications. |
| [12]                | Tween 80.                         | ii. Mix lemon oil, surfactant (Tween 80), and buffer |                                                          |
|                     |                                  | solution (10 mM phosphate buffer, pH 7.0) then blend together using a high-speed stirrer for 2 min at room temperature. |                                                          |
|                     |                                  | iii. Microfluidized using processor at 9000 psi. |                                                          |
| Type of emulsions/Literature Sources | Application/Dispersion materials | Preparation methods | Effect in physicochemical stability of bio-active compounds |
|-------------------------------------|----------------------------------|---------------------|----------------------------------------------------------|
| **Microemulsion** <sup>[13]</sup> | Dispersion of chitosan solution and microemulsions containing cinnamon bark oil and soybean oil to produce transparent antimicrobial films. | i. Add the oil and the polar phase into a closed container and mix them by hand-shaking until a fixed transparent appeared.  
ii. Combine chitosan stock and microemulsion for further antimicrobial films procedure. | • Cooperation of microemulsion improved microbiological safety and film transparency.  
  • Combination of chitosan stock with microemulsion slightly increased droplet size from less than 30 nm of microemulsions to larger than 88 nm.  
  • Transparent and low opacity antimicrobial films produced.  
  • Large zones of inhibition against foodborne pathogens observed. |
| **Microemulsion** <sup>[14]</sup> | Heat-treated whey protein isolate (WPI), date palm pit aqueous extract powder, glucono-d-lactone (GDL) and calcium chloride was microemulsified in a mixture of sunflower oil and sorbitan monooleate. It will be used to produce biopolymeric particles entrapping heat sensitive nutraceuticals at ambient conditions. The partial hydrolysis of GDL to gluconic acid resulted in cold gelation of whey proteins inside the nanodroplets of microemulsion which afterwards were precipitated by centrifugation. | Heat-treated WPI, extract free particles and extract-loaded capsules had mean sizes of 23, 304 and 230 nm, respectively.  
A controlled release character was suggested for nanoparticles based on the phenolics content measurement released from extract-loaded nanoparticles | |
| Type of emulsions | Application/Dispersion materials | Preparation methods | Effect in physicochemical stability of bio-active compounds |
|------------------|----------------------------------|---------------------|----------------------------------------------------------|
| Nanoemulsion     | Astaxanthin nanodispersions formation. The emulsifiers used were polysorbates (Polysorbate 20, Polysorbate 40, Polysorbate 60 and Polysorbate 80) and sucrose esters of fatty acids (sucrose laurate, palmitate, stearate and oleate). | i. Nanodispersion developed through emulsification-evaporation method.  
ii. Prepare solvent-in-water emulsion and then its conversion into nanodispersion by removing the solvent.  
iii. Produce aqueous phase by dissolving all emulsifiers in deionized water (1% w/w) containing sodium azide (0.02% w/w) under magnetic stirring.  
iv. Produce organic phase by dissolving astaxanthin (1% w/w) in dichloromethane.  
v. Mix and homogenized both aqueous and organic phase using a conventional homogenizer at 5,000 rpm for 5 min.  
vi. The initial emulsion produced was then passed through a high-pressure homogenizer (APV, Crawley, UK) at 50 MPa for two cycles.  
vii. Run rotary evaporation to separate the solvent from the fine emulsion at 250 Pa and 47 °C.  
viii. The formation of astaxanthin particles occurs by diffusion of the organic phase into the aqueous phase and evaporation at the air/water interface. | • The mean particle diameters of the nanodispersions ranged from 70 nm to 150 nm.  
• Higher emulsifier hydrophilicity and less carbon number of the fatty acid in the emulsifier structure reduced the particle structure.  
• Astaxanthin nanodispersions with the smallest particle diameters were produced with Polysorbate 20 and sucrose laurate among the polysorbates and the sucrose esters, respectively.  
• Most degradation of astaxanthin observed from Polysorbate 80- and sucrose oleate-stabilized nanodispersions |
| Type of emulsions/ Literature Sources | Application/Dispersion materials | Preparation methods | Effect in physicochemical stability of bio-active compounds |
|--------------------------------------|----------------------------------|---------------------|---------------------------------------------------------|
| Microemulsion [17]                   | Microemulsion of thymol using non-ionic (Tween 80), cationic (CTAB), and anionic (SDS) surfactants. | i. Mix surfactant–oil phase with a predetermined weight ratio of oil phase (ethanol + thymol) to surfactant (Tween 80, CTAB, or SDS) in glass vials, seal and store at 25 °C. ii. Titrate water into the oil-surfactant mixtures with vigorous shaking on a stirrer plate. The prepared samples were kept at 25 °C for equilibration for at least 24 h before examination. iii. Vortex the sample, remained homogenous-transparent sample categorized as microemulsions. | *Regarding to differential scanning calorimetry analysis result, microemulsions gradually inverted from the water-in-oil to the bicontinuous and finally to the oil-in-water microstructures upon dilution line at 90/10 surfactant/oil ratio. *Rod-like micelles turned to spherical micelles in the O/W region observed through rheological analysis. *Transition of w/o microemulsions into bicontinuous and o/w structures decreased the antimicrobial activity against Escherichia coli and Staphylococcus aureus while the DPPH scavenging activity increased. |
| Nanoemulsion [18]                    | Salvia multicaulis essential oil (containing monoterpene hydrocarbons (58.01%) and oxygenated monoterpenes (27.63%)) nanoemulsion using tween 80 and span 80 as surfactants, and water mixture by using high intensity ultrasound. | Nanoemulsion produced through high intensity ultrasound method. | *Compared to oil-free nanoemulsion, essential oil loaded nanoemulsion showed better antimicrobial activities against tested microbes. *The size, distribution of particle size and encapsulation efficiency were 89.45%, 0.38% and 69.9%, respectively, and the produced nanoemulsions were stable for 60 days. *All concentrations of the Salvia multicaulis nanoemulsion showed positive antioxidant activity. |
| Microemulsion [19]                   | Food-grade microemulsion of b-carotene using Tween-80 as surfactant. | i. Mix appropriate amounts of long chain triglycerides (soybean oil) or medium chain triglycerides, phosphate buffer, and surfactant (Tween 20, 40 or 80) using vortex mixer in vials at room temperature. | *Transparent b-carotene-encapsulated O/W microemulsions in the particle size range of 12–100 nm can be form using combination of short chain monoglycerides with Tween 80. *The microemulsion region of a ternary phase diagram containing short chain monoglycerides was larger than for di- and
ii. Equilibrate all samples at room temperature for at least 24 h before evaluation and re-examined after one week.

| Type of emulsions/ Literature Sources | Application/Dispersion materials | Preparation methods                                                                 | Effect in physicochemical stability of bio-active compounds |
|---------------------------------------|----------------------------------|--------------------------------------------------------------------------------------|-------------------------------------------------------------|
| Microemulsion                         | Ability comparison between two natural small molecule surfactants-quillaja saponin and soy lecithin to stabilize oil-in-water emulsions. | Emulsions containing 10% medium chain triglyceride (MCT) oil stabilized by 0.5-2.5% quillaja saponin or 1-5% soy lecithin were fabricated using high pressure microfluidization. | • The mean droplet diameter decreased with increasing emulsifier concentration
• Both emulsifiers led to the formation of oil droplets with a high negative charge ($\gamma = -45$ to $-70$ mV), thereby generating a strong electrostatic repulsion that helped protect them against aggregation.
• The emulsions remained physically stable when added to an acidic hot coffee solution (85 °C), with no visible phase separation or increase in particle size.
• This study provides insight into the potential of two natural emulsifiers to form stable emulsions suitable for application in coffee creamers. |
| Nanoemulsion                          | Improving shelf-life of low-fat cut cheese through development of nanoemulsion-based edible coatings containing oregano essential oil (OEO) (1.5-2.5% w/w), as natural antimicrobial compound, with combination of sodium alginate (2.0% w/w), mandarin fiber (0.5% w/w) and Tween 80 (2.5% w/w). | i. Prepare aqueous phase by solving sodium alginate in ultrapure water at 70 °C for 3 h.
ii. Add mandarin fiber to alginate solution and mix them using a laboratory high-shear homogenizer at 9600 rpm for 3 min.
iii. Add lipid phase consisted of the mixture of OEO and Tween 80 at room temperature to the aqueous phase, blend with the high-shear homogenizer at 11,000 rpm for 2 min, leading to coarse emulsions.
iv. Form nanoemulsion by passing the coarse emulsion through a microfluidizer at 150 MPa for 5 cycles. | • Coatings with at least 2.0% (w/w) OEO decreased Staphylococcus aureus population after 15 days.
• Coated-cheese pieces containing 2.5% (w/w) OEO inhibited psychrophilic bacteria or molds and yeasts growth during 6 or 24 days of storage, respectively. |
| Type of emulsions/ Literature Sources | Application/Dispersion materials | Preparation methods | Effect in physicochemical stability of bio-active compounds |
|---------------------------------------|----------------------------------|---------------------|----------------------------------------------------------|
| Microemulsion [22]                    | Production of active films from cellulose acetate by incorporation of pink pepper EO, evaluating this action by diffusion in solid medium (agar), dispersion in liquid medium (broth), volatilization (micro atmosphere), and in situ (sliced mozzarella cheese) against Staphylococcus aureus, Listeria monocytogenes, Escherichia coli and Salmonella Typhimurium. | The films were produced by casting technique. | - The concentrations of 2, 4 and 6% of EO added to the films made them active against L. monocytogenes and S. aureus in all evaluated media. - The in-situ tests demonstrated that the affinity between nonpolar molecules of EO and the lipid components of cheese allows the EO of the polymer to migrate to food, indicating favorable characteristics for its use as active packaging, by direct contact. |
| Nanoemulsion [23]                     | Development of sesame oil blended eugenol-loaded nanoemulsion using non-ionic surfactant (Tween20/Tween80) and water as a sources of antimicrobial for food preservation. | Ultrasonic cavitation method | - Lower droplet size (less than 13 nm) and higher stability obtained from sesame oil blended eugenol-loaded nanoemulsion compared to nanoemulsion without sesame oil. - Tween80 was more effective in reducing droplet size when compared to that of Tween20 |
| Nanoemulsion [24]                     | Nanoemulsion using whey protein-maltodextrin conjugate with propylene glycol as a co-surfactant. Preparation of stable and translucent thymol nanoemulsions using gelatin and lecithin in cantaloupe juice. Preparation of high emulsifying capacity of nanoemulsion using combination of sodium caseinate (NaCas) and lecithin. | High pressure homogenization method | - Compared to free thymol, the thymol-loaded nanoemulsions were consistently more efficient in inhibition of Listeria monocytogenes and Escherichia coli O157:H7 in milk and cantaloupe juice. - Nanoemulsions prepared with GRAS emulsifiers have great potential for use as novel antimicrobial preservatives to improve food safety. |
| Nanoemulsion   | Preparation of antimicrobial delivery system by encapsulating D-limonene into an organogel-based nanoemulsion using tween-80 as a ternary food-grade surfactant for food preservative application. | High pressure homogenization method | • Droplet size (d ± 36 nm) with better stability obtained.  
• Results from the antimicrobial activity have shown the encapsulation of D-limonene (4% w/w) into the organogel-based nanoemulsion contributed to the increase of its antimicrobial activity. |
|---------------|-------------------------------------------------------------------------------------------------------------------|-----------------------------------|---------------------------------------------------------------------------------|
| Nanoemulsion  | Nanoemulsion (NE) preparation using orange oil, Tween 80 (organic phase) and water (aqueous phase) to inhibit microbial spoilage in apple juice.                  | Sonication technique              | • Droplet size range from 20 to 30 nm obtained.  
• NE treated apple juice showed complete loss of viability even on dilution as compared to their controls. |
| [26]          | Development and characterization capsaicin-loaded nanoemulsions stabilized with natural biopolymer such as alginate and chitosan for use as a functional ingredient delivery system of functional food. | Self-assembly emulsification methods | • The particle sizes obtained were 20 nm or lower.  
• In conclusion, the double-layer nanoemulsions incorporated with alginate and chitosan can be expected to improve the stability of nanoemulsion. |
| Nanoemulsion  | Preparation of olive oil w/o nanoemulsions using non-ionic surfactants (Tween 20, Span 20) without the addition of a co-surfactant.                          | High pressure homogenization method | • The incorporation of olive oil endogenous compounds resulted into more stable emulsions.  
• In particular, gallic acid was proven to be the most efficient compound since it enhanced the emulsion properties prolonging simultaneously the emulsions’ stability. |
| [28]          | Dispersion of different olive oil endogenous compounds with an amphiphilic character (gallic acid, apigenin, quercetin, and transcinnamic acid) into the aqueous phase. |                                    |                                                                                       |
| Nanoemulsion  | Preparation of D-limonene in water nanoemulsion using mixed surfactants of sorbitane trioleate and polyoxyethylene (20) oleyl ether.                      | Ultrasound emulsification method   | • Investigation using response surface methodology revealed that 10% D-limonene nanoemulsions formed at S0 ratio (D-limonene concentration to mixed surfactant concentration) 0.6–0.7 and applied power 18W for 120s had droplet size below 100 nm. |
| [29]          |                                                                                                                  |                                    |                                                                                       |
| Type of emulsions/ Literature Sources | Application/Dispersion materials | Preparation methods | Effect in physicochemical stability of bio-active compounds |
|--------------------------------------|----------------------------------|---------------------|-----------------------------------------------------------|
| Nanoemulsion [30]                    | Encapsulation of a terpenes mixture and D-limonene into nanoemulsions based on food-grade ingredients for preservation of fruit juice. | High pressure homogenization at 300 MPa. | - Higher antimicrobial activity of the nanoencapsulated compounds allowed, least antimicrobial concentrations requirement for a bactericidal action with a minimal alteration of the organoleptic properties of the juice. |
| Nanoemulsion [31]                    | Enhancement of antimicrobials activities through encapsulation of carvacrol, limonene and cinnamaldehyde in the sunflower oil droplets of nanoemulsions stabilized by different emulsifiers like lecithin, pea proteins sugar ester and a combination of Tween 20 and glycerol monooleate. | High pressure homogenization method | - In particular, the antimicrobial activity was correlated to the concentration in the aqueous phase of the active molecules. |
| Nanoemulsion [32]                    | The purpose of this study was to investigate the antimicrobial activity of unilamellar nano vesicles (liposome) containing D-limonene against selected fruit rotting fungi (Botrytis cinerea and Penicillium chrysogenum) and food borne illness causing bacteria (Escherichia coli and Listeria monocytogenes). Further evaluation on the extended shelf life and enhanced food safety of blueberries treated with D-limonene and liposomes. Liposomal nanoparticles were created by thin lipid film hydration. | Sonication technique | - High stability with mean liposome radius 100.2-3.1 nm.  
- The in vivo study of liposome coatings on blueberries also revealed protection against microbial growth even after nine weeks of storage at 4 °C with liposomes reducing blueberry spoilage by more than 60% at the end of nine weeks.  
- The results of this study can benefit the produce industry through both enhancement of food safety and extending the shelf life of blueberries, further highlighting the commercial applications of liposomes. |
5. Conclusion
Microemulsion technology has been applied on laundry, cosmetic, as well as various food-types industries. It has been well-known delivery system according to their effectiveness and high stability when combined with another components. With microemulsion system, lipophilic water-insoluble component and hydrophilic water-soluble components could be well-solubilized in water-based substances as well as oil-based substances respectively. It will lead to the development of fine combination between two immiscible substances that complemented each other. For safety purpose and future awareness, replacement of these ternary food grade by suitable natural emulsifier ingredients is highly recommended.

6. References

[1] Shannon P, Callender, Jessica A. Mathews, Katherine K, and Shawn D. Wettig 2017, Review on Microemulsion utility in pharmaceuticals: Implications for multi-drug delivery. International Journal of Pharmaceutics, 526 425-442. DOI:10.1016/j.ijpharm.2017.05.005.

[2] Azhar Y K, Sushama T, Zeenat I, Farhan J A, and Roop K K 2006, Multiple Emulsions: An Overview. Current Drug Delivery, 3 429 - 443. DOI:10.2174/156720106778559056

[3] Sushama Talegaonkar, Adnan Azeem, Farhan J. Ahmad, Roop K. Khar, Shadab A. Pathan, Zeenat I, and Khan 2008, Microemulsions: A Novel Approach to Enhanced Drug Delivery. Recent Patents on Drug Delivery & Formulation, 2 238-257. DOI:10.2174/187221108786241679

[4] Lawrence M J, Rees G D 2000, Microemulsion-based media as novel drug delivery systems. Advanced Drug Delivery Reviews, 45 89-121

[5] John F and Harginder S 2006, Microemulsions: a potential delivery system for bioactives in food, Critical Reviews in Food Science and Nutrition, 46 221-327. DOI: 10.1080/10408690590956710

[6] Ghosh P K, Murthy R S R 2006, Microemulsions: A Potential Drug Delivery System. Current Drug Delivery, 3 167 - 180, DOI:10.2174/156720106776359168

[7] Jadhav K R, Shaikh I M, Ambade K W, Kadam V J 2006, Applications of Microemulsion Based Drug Delivery System. Current Drug Delivery, 3 267 - 273, DOI:10.2174/156720106777731118

[8] Huaiqiong C and Qixin Z 2015, Thermal and UV stability of b-carotene dissolved in peppermint oil microemulsified by sunflower lecithin and Tween 20 blend. Food Chemistry, 174 630–636. DOI: 10.1016/j.foodchem.2014.11.116

[9] Ariviani S, Anggrahini S, Naruki S and Raharjo S 2015, Characterization and chemical stability evaluation of β-carotene microemulsions prepared by spontaneous emulsification method using VCO and palm oil as oil phase. International Food Research Journal, 22 2432-2439, 2015.

[10] Maria D C, Evgenia M, Anan Y, Aristotelis X, Vassiliki P 2015, Formulation and characterization of food-grade microemulsions as carriers of natural phenolic antioxidants. Colloids and Surfaces A: Physicochem. Engineering Aspects, 483 130–136. DOI: 10.1016/j.colsurfa.2015.03.060

[11] Duan X, Li M, Ma H, Xu X, Jin Z and Liu X 2016, Physicochemical properties and antioxidant potential of phosvitin–resveratrol complexes in emulsion system. Food Chemistry, 206 102–109. DOI: 10.1016/j.foodchem.2016.03.055

[12] McClements, Jiajia R and David J 2012, Food-grade microemulsions and nanoemulsions: Role of oil phase composition on formation and stability. Food Hydrocolloids, 29 326-334
[13] Qiumin M, Yue Z, Faith C, P. Michael D, Svetlana Z and Qixin Z 2016, Physical, mechanical, and antimicrobial properties of chitosan films with microemulsions of cinnamon bark oil and soybean oil. *Food Hydrocolloids*, 52 533-542. DOI: 10.1016/j.foodhyd.2015.07.036

[14] Saeed S, Ashkan M and Mohamadsaeed Y 2014, Microemulsifications cold gelation of whey proteins for nanoencapsulation of date palm pit extract. *Food Hydrocolloids*, 35 590-596. DOI:10.1016/j.foodhyd.2013.07.021

[15] Jing T, Qin Z, Fang Q, Mingfu W, Jie C, and Zong P Z 2017, Preparation of steppogenin and ascorbic acid, vitamin E, butylated hydroxytoluene oil-in-water microemulsions: Characterization, stability, and antibrowning effects for fresh apple juice. *Journal of Food Chemistry*, 224 11-18

[16] Navideh A, Chin P T 2013, Physico-chemical stability of astaxanthin nanodispersions prepared with polysaccharides as stabilizing agents. *International Journal of Food Sciences and Nutrition*, 64 744-748

[17] Deng L, Taxipalati M, Sun P, Que F, and Zhang H 2015, Phase behavior, microstructural transition, antimicrobial and antioxidant activities of a water-dilutable thymol microemulsion. *Colloids and Surfaces B: Biointerfaces*, 136 859–866

[18] Sasan G, Nayyer K, Samira F, Manouchehr N, Saman G, Vahid J, Mahmoud S K, and Hossein S K 2017, Application of Salvia multiflora essential oil-containing nanoemulsion against food-borne pathogens. *Journal of Food Bioscience*, 19 128–133

[19] Roohinejad S, Oey I, Wen J, Lee S J, Everett D W, and Burritt D J 2015, Formulation of oil-in-water b-carotene microemulsions: Effect of oil type and fatty acid chain length. *Food Chemistry*, 174 270–278

[20] Cheryl C, Alexander S, Philippe R, Eric A D, and David J M 2017, Formulation of food emulsions using natural emulsifiers: Utilization of quillaja saponin and soy lecithin to fabricate liquid coffee whiteners. *Journal of Food Engineering*, 209 1-11, 2017.

[21] Artiga, María, Fani, Alejandra and Martin-Belloso, Olga. Improving the shelf life of low-fat cut cheese using nanoemulsion-based edible coatings containing oregano essential oil and mandarin fiber. *Food Control*, 76 1-12. DOI: 10.1016/j.foodcont.2017.01.001

[22] Guilherme S D, Graciele D F, Claudio E S C, Juliana L M, Wladimir P S, Ângela M F 2017, Essential oil from pink pepper as an antimicrobial component in cellulose acetate film: Potential for application as active packaging for sliced cheese, *LWT - Food Science and Technology*, 81 1-17. DOI:10.1016/j.lwt.2017.04.002

[23] Ghosh V, Mukherjee A, Chandrasekaran N 2014, Eugenol-loaded antimicrobial nanoemulsion preserves fruit juice against, microbial spoilage, *Colloids and Surfaces B: Biointerfaces*, 114 392–397. DOI: 10.1016/j.colsurfb.2013.10.034

[24] Xue J, Xue, Jia 2015, Essential Oil Nanoemulsions Prepared with Natural Emulsifiers for Improved Food Safety. PhD diss., University of Tennessee.https://trace.tennessee.edu/utk_graddiss/3381

[25] Mohamed R Z, Hao L, and Qipeng Y 2015, Improving the antimicrobial activity of D-limonene using a novel organogel-based nanoemulsion. *Food Control*, 50:554-559

[26] Saranya S, Sanjay S, Amitava M, and Chandrasekaran N 2015, Nanoemulsion of orange oil with non ionic surfactant produced emulsion using ultrasonication technique: evaluating against food spoilage yeast. *Applied Nanoscience*, 6 113–120. https://doi.org/10.1007/s13204-015-0412-z
[27] Ae-Jin C, Chul-Jin K, Yong-Jin C, Jae-Kwan H, and Chong-Tai K 2011, Characterization of Capsaicin-Loaded Nanoemulsions Stabilized with Alginate and Chitosan by Self-assembly, Food Bioprocess Technology. 4 1119–1126. https://doi.org/10.1007/s11947-011-0568-9

[28] Vasiliki P and Constantina T 2016, Study of the Emulsifying Ability of Olive Oil Endogenous Compounds in Co-surfactant Free Olive Oil w/o Nanoemulsions with Food Grade Non-ionic Surfactants. Food Bioprocess Technology, 91 1-10. DOI:10.1007/s11746-013-2356-3

[29] Po-Hsien L and Been-Huang C 2012, Process optimization and stability of D-limonene-in-water nanoemulsions prepared by ultrasonic emulsification using response surface methodology. Ultrasonics Sonochemistry. 19 192–197.DOI:10.1016/j.ultsonch.2011.05.017

[30] Francesco D, Marianna A, Mariarenata S, and Giovanna F 2011, Nanoencapsulation of essential oils to enhance their antimicrobial activity in foods, LWT - Food Science and Technology. 44 1908-1914

[31] Francesco D, Mariann A, Mariarosaria V, and Giovanna F 2012, Design of nanoemulsion-based delivery systems of natural antimicrobials: Effect of the emulsifier. Journal of Biotechnology, 159 342–350. https://doi.org/10.1016/j.jbiotec.2011.07.001

[32] Arosha L U, Nathalie B, Punit K, Derek J F, and Ruplal C 2017, Antimicrobial efficacy of liposomes containing D-limonene and its effect on the storage life of blueberries Article in press, Postharvest Biology and Technology, 128 130-137. https://doi.org/10.1016/j.postharvbio.2017.02.007

Acknowledgments
Authors wishing to acknowledge financial support from the Office of Innovation, Commercialization and Consultancy Management (ICC) through the TIER1 funding vote no. H256 and GPPS funding vote no. H052 provided by the Universiti Tun Hussein Onn Malaysia (UTHM) in assistance with the Malaysia Government is gratefully acknowledged.