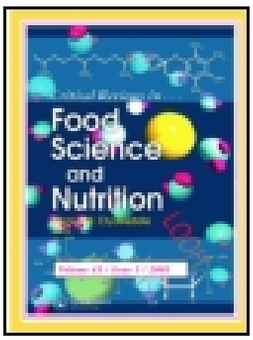

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Food Emulsions as Delivery Systems for Flavor Compounds — A Review

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Abstract

Food flavor is an important attribute of quality food, and it largely determines consumer food preference. Many food products exist as emulsions or experience emulsification during processing, and therefore, a good understanding of flavor release from emulsions is essential to design food with desirable flavor characteristics. Emulsions are biphasic systems, where flavor compounds are partitioning into different phases, and the releases can be modulated through different ways. Emulsion ingredients, such as oils, emulsifiers, thickening agents, can interact with flavor compounds, thus modifying the thermodynamic behaviour of flavor compounds.

Emulsion structures, including droplet size and size distribution, viscosity, interface thickness, etc., can influence flavor component partition and their diffusion in the emulsions, resulting in different release kinetics. When emulsions are consumed in the mouth, both emulsion ingredients and structures undergo significant changes, resulting in different flavor perception. Special design of emulsion structures in the water phase, oil phase and interface, provides emulsions with great potential as delivery system to control flavor release in wider applications. This review provides an overview of the current understanding of flavor release from emulsions, and how emulsions can behave as delivery system for flavor compounds to better design novel food products with enhanced sensorial and nutritional attributes.

Keywords

food flavor, emulsion, delivery system, oil, controlled release

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1. Introduction

There is growing interest in functional foods with low fat, low sugar, low salt, or bioactives-enriched, to develop healthier diets for human wellbeing. In the meantime, consumers ask for food with desirable organoleptic properties, particularly texture and flavor profile. It is a big challenge for academic and industrial researchers to design healthy food products without sacrificing food flavor, as flavor release is not only influenced by food ingredients but also food structures (Druaux and Voilley, 1997). Either faster release or slower release than normal release may result in unfavourable flavor perception. Therefore, researchers have been working on designing delivery systems for flavor compounds to better control their release when incorporated in foods, and subjected to different environmental stresses encountered during product processing, storage and mastication in the oral cavity.

Emulsions consist of two immiscible phases, one of which is dispersed in the other as small droplets; these are known as dispersed phase and continuous phase, respectively (McClements, 2005). Functional food ingredients can be incorporated into the dispersed droplets, and thus isolated from the external environment by the continuous phase. Many studies have shown that emulsions are effective delivery systems for functional food ingredients (bioactive lipids, anti-oxidants, flavor, etc.) to protect them from degradation (mechanical, chemical, enzymatic), to disperse them in aqueous media, to control their release and finally to improve their

bioavailability in the human gastro-intestinal tract (Velikow and Pelan, 2008; McClements, 2010). An important characteristic of an emulsion is that the structures in water phase, oil phase and interface can be designed to meet special requirements, and the release of the compounds incorporated can then be modified (Mao et al., 2015; McClements and Li, 2010). Flavor release from emulsions includes the partitioning and mass transfer of the flavor molecules among oil phase, interface, water phase, and finally headspace (Taylor, 1998). Changes in headspace concentration and release rate could affect the overall flavor perception. The successful development of delivery systems with controlled flavor release depends on a good understanding of the effects of emulsion properties (e.g., droplet size and distribution, viscosity), and of environmental stresses affecting flavor release, as well as the interactions between flavor compounds and emulsion components. Recent work suggested the possibility to modulate flavor release, including release rate and intensity, by modifying emulsion microstructures through food engineering techniques (Ghosh et al., 2006; Benjamin et al., 2012a; Mao et al., 2014).

The objective of this article is to give an updated overview of recent advances in emulsion-based delivery systems for food flavors. A brief summary is firstly made on the theories developed to describe flavor release from emulsions and our understanding about flavor release in the mouth. More emphasis is made on factors affecting flavor release, i.e., different emulsion ingredients and emulsion properties, and how controlled flavor release can be achieved by appropriate

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modification of all structural elements of the food dispersion. In the last section, novel structured emulsions with the potential to better deliver flavors in more complex systems are introduced. It should be noted, however, that the flavors discussed in this review are smell-related compounds (volatiles), whereas taste-related compounds are not included.

2 Physical Chemistry of Flavor Release

Volatile flavors are perceived when they are in contact with receptors in the nose (olfactory epithelium) either orthonasally by directly sniffing foods or retronasally by delivering of volatiles during mastication and/or swallowing. Before flavor perception is realized, flavor compounds have to move out from food matrix, and experience immigration in oral cavity (and/or nasal cavity).

2.1 Flavor Release from Emulsions

Flavor release is mainly controlled by two factors, the volatility of flavor compounds (thermodynamic factor) and the resistance to mass transfer from the emulsion to the air phase (kinetic factor) (de Roos, 2000). The thermodynamic factor determines the retention or partition of flavors in the matrix at equilibrium, and the kinetic factor mainly affects the release rate of flavors from food. Flavor release from an O/W emulsion can be simplified into four steps: (1) flavor movement inside the oil droplets; (2) movement over the oil-water interface; (3) movement within the aqueous phase; (4) movement across the air-emulsion interface (Figure 1).

Theoretically, each step affects the release rate and contributes to the headspace concentration. In reality, only one or two steps can dominate the release.

2.1.1 Partition Equilibrium in Different Phases

Static headspace analysis is widely applied to evaluate flavor release, and it is based on the theory of partition equilibrium. In a closed system, flavor compounds dissolved in an emulsion can reach partition equilibrium in different phases, and the partition coefficient is dependent on the affinity of flavor compounds for each phase. Therefore, for a simple O/W emulsion, the overall distribution of flavor compounds between emulsion matrix and its headspace can be expressed as (Buttery et al., 1973):

$$\frac{1}{K_{GE}} = \frac{\varnothing_D}{K_{GD}} + \frac{\varnothing_C}{K_{GC}} \qquad (1)$$

Where K_{GE} is the partition coefficient of a flavor between air (headspace) and emulsion; ϕ_D is the mass fraction of dispersed phase; ϕ_C is the mass fraction of continuous phase ($\phi_D + \phi_C = 1$); K_{GC} is the partition coefficient between air and continuous phase (aqueous emulsifier solution) and K_{GD} is the partition coefficient between air and dispersed phase. Several studies showed that this model could give a good prediction of headspace-emulsion partition coefficient when the interfacial binding was insignificant (Guyot et al., 1996; Ghosh et al., 2007). On the other hand, the difference in the calculated and experimental K_{GE} values can be regarded as proof of interfacial binding (Karaiskou et al., 2008; Meynier et al., 2005).

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In many emulsions, particularly the ones stabilized by biopolymers, flavor binding to interfacial components has been widely observed, and it plays a significant role in flavor partition (Guichard, 2002). For reversible binding, a modified model of equation (1) describing flavor partition was proposed by McClements (2005):

$$\frac{1}{K_{GE}} = \frac{\varnothing_D}{K_{GD}} + \frac{\varnothing_C}{K_{GC}} + \frac{A_S K^*_{IC}}{K_{GC}} \qquad (2)$$

Where A_S is the interfacial area per unit volume of an emulsion ($A_S = 6\phi/d_{32}$, d_{32} is the volume-surface mean droplet diameter); K^*_{IC} is the interfacial binding coefficient ($K^*_{IC} = \Gamma_I/C_C$, Γ_I is the mass of flavor compound per unit of interfacial area; C_C is the flavor compound concentration in the continuous phase). From Equation (2), it is observed that K_{GE} is droplet size-dependent. When droplet size is large enough, A_S will be infinitesimal. Then interfacial binding can be neglected, and Equation (2) is equal to Equation (1).

For irreversible binding, McClements (2005) proposed a different model:

$$\frac{1}{K^e_{GE}} = \left(\frac{C_E}{C_E - A_S\Gamma_I}\right)\left(\frac{\varnothing_D}{K_{GD}} + \frac{\varnothing_C}{K_{GC}}\right) \qquad (3)$$

Where C_E is flavor concentration added to the emulsion, and Γ_I is the mass of flavor compounds irreversibly bound to the interface per unit of interfacial area. It should be noted that Equation (2) and Equation (3) do not involve flavor binding in the continuous phase or dispersed phase, e.g., flavor solubilisation in emulsifier micelles, gum binding. As a result, the predicted values of K_{GE} could be higher than the experimental values.

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2.1.2 Flavor Release from Real Food

The eating processes usually finish in a short time, and flavor release hardly reaches equilibrium. Therefore, the perception of volatile flavor mainly depends on the initial dynamic release. The theory of dynamic release reveals more information about the process of flavor release, as more parameters are considered, which on the other hand results in the application of more complex models to describe the process. The following equation proposed by Harrison and Hills (1997) is an example of the models used to predict headspace concentration at a given time (t):

$$C_g(t) = \frac{C_{tf}(0)}{\frac{1+K_bC_b}{K_{ge}} + \left(\frac{v_g}{v_e}\right)\left[1-\exp\left\{-\frac{A_{ge}h_D}{v_g}\left(\frac{1}{K_{ge}}+\frac{v_g}{v_e}\frac{1}{1+K_bC_b}\right)t\right\}\right]} \quad (4)$$

where $C_{tf}(0)$ is the initial flavor concentration (mg cm^{-3}) in the emulsion, K_b is the flavor binding affinity to biopolymers (including some emulsifiers) (M^{-1}), C_b is the biopolymer concentration in the emulsion (M), v_g and v_e are the headspace and emulsion volumes (cm^3), and A_{ge} is the gas/emulsion surface (cm^2). The mass transfer coefficient h_D (cm s^{-1}) and the gas/emulsion partition coefficient K_{ge} can be derived from the above equation at the initial release rate ($t \rightarrow 0$) and at the equilibrium headspace concentration ($t \rightarrow$ infinite), respectively. This model follows the penetration theory, which was developed to describe flavor release from liquid samples. In this theory, flavor transfer through the gas-liquid interface is assumed to be rate-limiting, and is driven by molecular diffusion and eddy diffusion. The binding effect is taken into consideration

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in this model, and it has been successfully applied to predict flavor release from different liquid systems (Perreault et al., 2010; Benjamin et al., 2012a).

Lian et al. (2004) have developed another model which took two rate-limiting steps into consideration: one was the flavor transfer from droplets to the continuous phase; the other one was the flavor transfer from continuous phase to the headspace. Actually, for emulsions with smaller droplet size, where flavor release is governed by the mass transfer across the air-emulsion interface, this model is similar to the model developed by Harrison and Hill (1997). Nevertheless, for emulsions with larger droplet size, the Lian et al.'s model suggested that the transfer across droplet interface becomes rate-limiting, as the larger droplets can offer a significant barrier to diffusion. In Lian et al.'s study on gelled emulsions, the experimental data of four ketones and one ester fitted the model well (Lian et al., 2004).

2.2 Flavor Release during Eating

Food in the mouth experience mastication, salivation, bolus formation and finally swallowing. Each step can have a big influence on flavor release and flavor perception. Liquid foods normally stay in the mouth for several seconds before swallowing, so only one or two of the above processes have a significant effect on flavor release. When liquids are kept in the mouth, a tight closure formed by the soft palate and the base of the tongue prevents flavor transfer from the oral cavity to the nasal cavity. There is also no flavor transfer during swallowing, as the airway from the trachea to the nose is closed at that stage (Bosman, 1980; Normand et al., 2004).

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As a result, flavor perception is only available after swallowing (Land, 1996). Studies suggested that the highest perception was achieved in the first expiration immediately after swallowing (Linforth and Taylor, 2000). These flavors originate from the air phase above liquid food in the mouth before swallowing, which are later transported to the throat during swallowing, and finally transferred to the nose in the first expiration. This process is called "swallow breath", which finishes in a short time (de Roos and Wolswinkel, 1994; Land, 1996). There is another process of flavor transfer after swallowing with much lower flavor concentration, which contributes to the persistence of flavor perception. Specifically, a thin film of liquid food will remain at the surface of the pharynx after swallowing, and the flavor in the film can be released and transported to the nasal cavity during exhalation. As there is a gradient in flavor concentration between the thin film and the exhaled air, this release can last for a short period of time (Buettner et al., 2002; Weel et al., 2004a; Boelrijk et al., 2006). Moreover, the little amount of liquid food remaining in the mouth (adsorbed to the mucosa) after the first swallowing may also contribute to the persistence of flavor perception (Harrison, 1998; Salles et al., 2011).

3 Emulsion Ingredients and Flavor Release

3.1 Oil Phase

Most food flavors are lipophilic, and oils play a more significant role in their release than any other emulsion ingredient, e.g., proteins (Innocente et al., 2014). Oils can act as flavor precursors, as solvents for flavor compounds, and as flavor release modulators. Change in oil property or oil

content, can lead to a significantly modified flavor release profile (de Roos, 1997), which is difficult to be recovered by adding fat replacers. However, consumers are highly cautious about the fat/oil content of food, as overconsumption of fat/oil can increase the risks of many diseases, e.g., obesity, cardiovascular diseases, cancers (Gastaldelli and Basta, 2010). Therefore, knowledge about the effects of oil on flavor release becomes essential for food scientists to properly design fat-reduced food.

3.1.1 Oil Property

Flavors in different oils generally have different release behaviours, due to their affinity for different oils. The chain length of fatty acids determines the polarity of oils, which is one of the main factors affecting the interactions between flavor compounds and oils. Most flavor compounds are lipophilic, and they have higher affinity for oils containing longer-chain fatty acids (Harvey et al., 1995). However, there was a study showing that some hydrophobic flavor compounds had higher release from the emulsion containing lipids with average carbon number (CN) of C14 or C16 than from the emulsion containing C9 lipids (Rabe et al., 2003a). Another study reported no difference in flavor release when changing the lipid type from milk fat (C16 and C18) to MCT (C6 and C8) in emulsions (Roberts et al., 2003a). It seemed that lipophilicity of different oils was not the only factor that influenced the affinity of flavors for the oils, or the affinity had already been so high that change in lipophilicity of the oils did not show any effect in the mentioned studies. Rabe et al. (2003a) suggested that the molarity of oil (droplet

concentration) in the system could give additional effect. They made emulsions containing the same molarity of C16 and C9, but differed in the mass fraction. These authors did not observe any significant difference in flavor release between the two emulsions. Roberts et al. (2003a) proposed that the saturation level of different oils could affect flavor partition: with stearine (a more saturated fat), flavor release was shown to be slower and of lower intensity than the release from olein (a more unsaturated fat). This result was in agreement with observations made by Welsh and Williams (1989), who found the oil/water partition coefficients of many flavors were lower in olive oil than in sunflower oil. However, Roudnitzky et al. (2003) found that the volatility of ethyl hexanoate in vegetable oils differing by their percentages of oleic (C18:1), linoleic (C18: 2), and linolenic (C18:3) decreased when the unsaturation level increased. The above inconsistent findings revealed that oils may affect flavor release through different mechanisms, depending on oil nature (composition and physical state) and flavor characteristics.

Lipids in emulsions can indeed be present at solid (i.e., fat), partial solid or liquid (i.e., oil) state, which can exhibit different effects on flavor release (Roberts et al., 2003a; Relkin et al., 2004). The general consensus is that flavor compounds can only partition into liquid oil, and the formation of solid oil could inhibit the migration of flavor compounds (Maier, 1975; McNulty and Karel, 1973). In an O/W emulsion, flavor compounds had higher headspace concentrations above a solid droplet emulsion than those above a liquid droplet emulsion (Relkin et al., 2004; Ghosh et al., 2006). Nevertheless, the initial sorption rates of flavor compounds were much

higher for the solid lipid, which was attributed to the adsorption of flavor compounds onto the surface of solid lipid particles (Ghosh et al., 2006). In a milk-based liquid emulsion, Roberts et al. (2003a) found that increase in the content of solid palm fat resulted in accelerated flavor release. However, when the melted palm fat was tested, there was no difference in flavor release from emulsions with different oils (Roberts et al., 2003a). In a more complicated situation, flavor compounds were dissolved in liquid oil before oil solidification, and flavor release was reported to slow down as oil crystallization went on (increase in solid fat content). This finding could be explained by the fact that flavor molecules were trapped within the solid particles, and the movement of flavors across the droplet surface was inhibited (Roberts et al., 2003a).

3.1.2 Oil Content

The oil content of food has been shown to affect not only the perceived intensity but also the temporal profile of flavors, as well as the release behaviour throughout storage. Oil reduction or even complete omission of an oil phase in food can lead to a drastic shift of the overall flavor profile. Fat-free products therefore often show an undesirable transient flavor burst, as the release is no longer mediated by a fat phase (Rabe et al., 2003a).

It has been well documented that a reduction in oil content will accelerate the release of lipophilic flavor compounds, and the headspace concentration of flavors above an emulsion with lower oil content is normally higher. In other words, to trigger the same flavor release, flavor compounds added to emulsions with lower oil content could be at lower level than that added to

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emulsions with higher oil content (Bayarri et al., 2006). Only a small portion of flavors (volatile) are hydrophilic compounds, and they have different release behaviours. Both *in vivo* and static headspace studies showed that the release of hydrophilic flavor compounds was not or even positively affected by changing oil content (Rabe et al., 2003a; Giroux et al., 2007; Frank et al., 2011). The effect of oil content on flavor release is so dominant that sometimes it makes the contribution of other ingredients undetectable. Roberts and Pollien (2000) found that the evidence of protein or lactose binding with lipophilic compounds was seen as the concentration of skim milk increased, but the binding effect was no longer detected after the addition of 1.3% lipid. A similar study found that in the presence of ≥ 1% oil in an oil (MCT)-water system, the bindings between flavor compounds and β-lactoglobulin were insignificant (Seuvre et al., 2000). Giroux et al. (2007) compared the effect of oil and protein content on flavor release from emulsions and found that increasing the oil content (from 1-8%) reduced flavor release eight times more than when protein (from 0.1 to 3.16%) was added to the system.

The oil content also plays an important role in flavor perception under oral conditions, but the role is not as strong as that under static condition. (Weel et al., 2004b; Roberts et al., 2003b; Doyen et al., 2001; Karaiskou et al., 2008). As some researchers suggested, the perceived flavor is from the "swallow breath" and the thin liquid layer formed on the throat surface, and the role of oil as flavor reservoir is not as significant as that in the bulk phase situation (Land, 1996; Linforth and Taylor, 2000). Moreover, flavor perception could be affected by the presence of

sweets and viscosity of different systems, and the role of flavor intensity may be weakened (Bayarri, et al., 2006).

Finally, a change in oil nature or oil content may influence flavor release indirectly by modifying the physicochemical properties of the emulsions concerned, for example, droplet size, viscosity, and emulsion stability (discussed in a later part).

3.2 Water Phase

The water phase generally makes up the majority fraction of an O/W emulsion, and it may contain many ingredients, e.g., emulsifiers, thickening agents, salts, minerals. Therefore, the water phase also contributes a lot to the overall perception of a food emulsion. All the ingredients may interact with flavor compounds, thereby modifying their release behaviours. However, as only a small part of volatile flavor compounds are hydrophilic, the water phase does not influence flavor release as much as the oil phase.

3.2.1 Emulsifiers

Emulsifiers play critical roles in the formation of emulsions, and they have a strong impact on most of emulsion properties. When flavor compounds are incorporated in emulsions, the emulsifiers can influence flavor release through different mechanisms. Among all of food emulsifiers, proteins are the most widely used and extensively investigated emulsifiers regarding flavor release. It is generally acknowledged that proteins in emulsions can slow flavor release, and the effects are dependent on protein and flavor types. Widder and Fisher (1996) observed

that sodium caseinate had the capacity to inhibit the release of esters, and Maier (1970) found casein and egg albumin could bind acetone and acetaldehyde. Dubois et al. (1996) reported an increase in retention of diacetyl and diallyl sulphide in a model cheese system with higher interfacial concentration of calcium caseinate. Some studies revealed that β-lactoglobulin in emulsions can modify the release behaviours of aldehydes, ketones and esters (Guichard, 2002). Charles et al. (2000) indicated that ethyl hexanoate exhibited significantly higher release from the emulsion containing α-lactalbumin than from the emulsion containing β-lactoglobulin. However, once the proteins were hydrolyzed by enzymes, the peptides had reduced capacity to retain flavor compounds (Wong et al., 2013).

The reduced flavor release in protein-stabilized emulsions is achieved mainly through two different mechanisms. First, proteins are likely to interact with flavor compounds, through reversible or irreversible interactions. In most cases, flavor compounds interact with proteins through hydrophobic binding or hydrogen bonding, both of which are reversible (Lubbers et al., 1998). Wu et al. (1999) proposed that the most probable binding site for these compounds was the hydrophobic pocket (the central calyx) within the protein structure. In the case of aldehydes, covalent irreversible binding with proteins was also reported (Gremli, 1974; O'Keefe et al., 1991), and these compounds are likely to be bound at the protein surface (Lübke et al., 2000). For a series of different milk proteins, the binding capacity to 2-nonanone followed the order: bovine serum albumin > β-lactoglobulin > α-lactalbumin > α_{s1}-casein > β-casein, and the

capacity of whey protein isolate (WPI) was stronger than that of sodium caseinate (Kühn et al., 2007). According to Harrison and Hills (1997), binding reduces the concentration of free flavors in the aqueous phase and, consequently, the flavors released into headspace. Second, proteins adsorbed at the interface can act as barriers to slow mass transfer of flavor molecules, leading to reduced release rates (Harvey et al., 1995). For example, the presence of β-lactoglobulin at a miglyol–water interface strengthened the resistance against the transfer of benzaldehyde across the lipid layer (Rogacheva et al., 1999). A similar result was found by Guichard and Langourieux (2000), who reported that the presence of interfacial β-lactoglobulin inhibited the transfer of hydrophobic flavor compounds from the oil droplets to water phase. Land (1978) further confirmed these findings, and it was indicated that the inclusion of a small amount of emulsifier in a non-emulsified oil-water system had no effect on the headspace concentration of dimethylsulfide, whereas in an emulsified system the headspace concentration was significantly decreased. Due to the large variations in physicochemical properties of the proteins, the interfaces formed by different proteins may have different thickness, porosity, compactness, etc. (Wilde et al., 2004; McClements, 2005), which could result in different barrier effect on flavor release. Benjamin et al. (2014) studied the effect of legume proteins (soy, pea, and lupin) on flavor release, in comparison with β-lactoglobulin. The findings suggested that the ability of legume proteins to interact with flavors was comparable with β- lactoglobulin for the affinity to

more hydrophobic compounds, and different legume proteins showed similar effect on flavor release.

In terms of some small molecular weight surfactants (e.g., tweens, spans), only limited studies have beeen performed concerning their effects on flavor release. Although no sufficient proof is available about the interactions between surfactants and flavor compounds, there was evidence showing that changes in surfactant concentration and type in the emulsion can significantly modify partition coefficients of many flavor compounds (van Ruth et al., 2002a). Also, the volatility of flavor compounds is reduced in the presence of a surface active compound than in water alone (Landy et al., 1996). On the other hand, when surfactants formed micelles/reverse micelles in the emulsions, they were able to incorporate flavor molecules and modify their release behaviours (Rabe et al., 2003a; Benjamin et al., 2014).

Finally, it is important to point out that emulsifiers play significant roles on emulsion properties, particularly on droplet size and emulsion stability, which may also show big influences on flavor release (discussed in a latter part).

3.2.2 Thickening Agents

Thickening agents are normally added to increase emulsion viscosity, so as to get desired emulsion stability and textural properties. Moreover, some thickeners can be used as fat replacers to formulate fat-reduced food (Conforti et al., 1996). The addition of thickening agents can also modify flavor release. This is because the thickening agents can create physical barriers inside

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emulsions against the mass transfer of flavor compounds (discussed in a later part). When the thickeners formed a gel network, flavors could be trapped inside and the release was inhibited (Guichard et al., 1991). Interactions between thickening agents and flavor compounds have been also observed, though the contribution to flavor retention was rather small (Boland et al., 2004; Karaiskou et al., 2008). Pectin could interact with flavor compounds through van der Waals attraction forces between the alkyl patch of the flavor molecules and the hydrophobic domains of pectin molecules (Maier, 1970). Moreover, hydrogen atoms in the undissociated carboxyl group of pectin could interact with unshared electron pairs of heteroatoms and oxygen atoms of flavor molecules via hydrogen bonding (Braudo et al., 2000). Hydroxypropyl methylcellulose (HPMC) was shown to bind allyl disulfide (Cook et al., 2003), and it could result in lower flavor perception (Ferry et al., 2006). In an emulsified edible film matrix, κ-carrageenan was suggested to bind ketone with its –OH group (Marcuzzo et al., 2010). However, κ-carrageenan in a whole milk system did not affect the headspace concentration nor the *in vivo* release of the flavors (González-Tomás et al., 2007). Additionally, the texture of the food created by a thickening agent can also affect flavor release (see sub-section 3.4.2).

3.2.3 Salts

People have long realized that the addition of salts can enhance flavor perception, and the phenomenon is termed “salting-out”. For hydrophilic flavor compounds, “salting-out” results from the reduction in the number of water molecules available to solubilise flavor compounds

(Nawar, 1971). For lipophilic flavor compounds, "salting-out" can lower their concentration in the water phase and drive them further to the oil phase and out into the gas phase. Therefore, higher salt concentration in emulsions can lead to a greater release of flavor compounds (Poll and Flink, 1984; Rabe et al., 2003b; Benjamin et al., 2012a; Bortnowska, 2012). A study on the effects of different salts in a protein stabilized system indicated that addition of NaCl and KCl had stronger salting out effect for methyl-butanal, hexanal, octanal, methional and pentanone than the addition of $MgCl_2$ and $CaCl_2$. It was proposed that salts affected the interactions between flavor compounds and proteins, and that NaCl and KCl can weaken the bindings to a higher level (Pérez-Juan et al., 2006). However, when flavor compounds were incorporated into oil droplets, "salting out" only gave slight influence on their partition. The presence of salts in an emulsion also fosters electrostatic screening effects, which can further modify the structure of the interfacial film, and result in modified interactions between flavor compounds and the interface. Some emulsions can even collapse (e.g., phase separation) after a certain period of storage in salted systems, and flavor compounds could move more freely from the dispersed phase to the continuous phase, leading to intensified flavor release. Salt-triggered flavor release is generally concentration-dependent. However, in many cases there is a critical concentration point above which a higher salt concentration does not further affect the flavour release process (Benjamin et al., 2012a).

4 Emulsion Properties and Flavor Release

4.1 Droplet Size

As discussed previously, droplet characteristics show large effects on the physicochemical properties of an emulsion. In an O/W emulsion, flavors have to move out from the oil droplets to the water phase before transferring to the air phase, and the transferring rate may be dependent on droplet size. Many studies have been carried out to understand the effect of droplet size on flavor release, but no consistent conclusion can be made. Smaller droplets have larger interfacial area, and the travel path from the droplet centre to the interface is shorter, both of which can induce faster mass transfer of flavor molecules. This hypothesis was confirmed by Benjamin et al. (2012a), who found that the release rates of flavor compounds in an aggregated emulsion were lower than those in a finely distributed emulsion. Other studies argued that smaller droplets with larger interfacial area can absorb more emulsifiers, which would result in slowed flavor release (van Ruth et al., 2002b) and reduced air-liquid partition coefficients (Meynier et al., 2005; van Ruth et al., 2002b).

However, the transfer of flavor compounds between the dispersed phase and continuous phase is generally thought to be very fast, especially when droplet size is reduced to micrometre or sub-micrometre range. Therefore, it may be difficult to find any significant difference in flavor release from two emulsions with different droplet size but within the same size range (Rabe et al., 2003a; Carey et al., 2002; Miettinen et al., 2002; Matsumiya et al., 2015).

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4.2 Rheological Behaviour

There is evidence that flavor perception can be suppressed by increasing the viscosity of a solution, though the magnitude of the suppression may vary among different flavors (Ferry et al., 2006). The Stokes–Einstein equation reveals that diffusion is negatively proportional to viscosity. Therefore, an increase in viscosity will reduce flavor diffusion in different phases. A slower flavor release was observed in emulsion systems containing pectin or gum arabic (Karaiskou et al., 2008; Druaux and Voilley, 1997). Baines and Morris (1987) reported that in order to produce a three-fold reduction in flavor perception using guar gum, it was required to increase the viscosity of the system by at least two-fold. In a gel-like viscous system, flavor release could be affected by the gel network structure created by gelling agents. Studies showed that increase in gel hardness can reduce flavor released to the headspace; i.e. firm gels made from carrageenan and gelatin could enhance flavor retention (Carr et al, 1996; Guniard and Marty, 1995). It has been also observed that flavor release was slower in the gels with higher gelatin concentration and that the release rate was dependent on the rate at which the gels collapsed after melting and chewing (Bakker et al., 1996). When different flavors were considered, hydrophobic flavor compounds were more likely to be affected. For different gelling agents, pectin and starch had relatively weaker effect on flavor release than gelatin, probably because of their less compact gel network structures (Boland et al., 2004). In a system containing Ca-alginate gels, flavor release was highly dependent on Ca^{2+} -alginate ratio. With higher Ca^{2+} concentration, the gel strength

was enhanced, and less flavor was released from the system (Baines, Morris, 1987). Hollowood et al (2002) revealed that the perceived strawberry flavor in Hydroxypropyl methylcellulose (HPMC) thickened solution was largely reduced when HPMC concentration was above a critical value (known as "coil overlap concentration"), at which HPMC starts to form an entangled network.

However, other studies failed to demonstrate any difference in flavor release on adjusting emulsion viscosity (Basaran et al, 1998; Siefarth et al, 2011). It was thus proposed that the pores of the biopolymer network were much larger than the size of the diffusing flavor molecules, and flavor molecules can travel through the entangled biopolymer chains without any barrier effect (Basaran et al., 1998). Another possible reason might be that the concentration of the thickener was below the coil overlap concentration in these systems (Siefarth et al., 2011). It was also argued that, when the headspace concentration of flavor is governed by the mass transfer at the air-liquid interface rather than that at the oil-water interface, the viscosity of the emulsion system can only exhibit a slight effect on flavor release (Roberts et al., 1996).

4.3 pH

Acids are common food ingredients, which besides other functions in composite food systemsthey are sometimes used as flavor enhancers. Acids in food not only influence taste, but also modify aroma release. In a soft drink system, a pH change from 5.0 to 3.0 or 4.0 (using citric acid) resulted in a significant increase of the release of esters (isopentyl acetate and ethyl

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hexanoate; Hansson et al., 2001). Nevertheless, when the pH was lowered to 2.0, with further addition of citric acid did not show any effect on the release of esters, menthone, and linalool. When the pH was regulated by a mixture of citric acid and sodium hydroxide, there was no difference in the release of esters, menthone and linalool among samples at pH 3.0, 4.0 and 5.0. The authors have attributed the results mainly to the chelating effect of the dissociated form of citric acid ($RCOO^-$);the more citric acid added to the solution, the greater the amount of dissociated citric acid would be available to interact with flavors, leading to lower headspace flavor concentration in the sample at pH 2.0. When both citric acid and sodium hydroxide were added, all the samples (pH 2.0, 3.0, 4.0, 5.0) had large amount of $RCOO^-$ (sample at pH 5.0 had the highest), and therefore the difference in flavor release could not be detected (Hansson et al., 2001).

A pH adjustment also affects the p*K* values of flavor compounds, which modifies the partition and release of flavors (Bennett, 1992). Moreover, the properties of emulsifiers (especially proteins) are highly dependent on pH. A change in pH can largely modify the interactions between emulsifiers and flavor compounds. For example, when pH was adjusted from 3.0 or 4.0 to 5.0, more than 30% of ethyl hexanoate, 2-decanone and 1-octanol were released to the headspace above emulsions stabilized by β-lactoglobulin (Benjamin et al., 2012a). In egg yolk or starch sodium octenylsuccinate stabilized emulsions, an increase of pH from 3.0 to 9.0 resulted in an enhanced retention of diacetyl, which was attributed to the strengthened interactions

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between diacetyl and the stabilizers through electrostatic attraction or hydrogen bonding at alkaline conditions (Bortnowska, 2012).

Jouenne and Crouzet (2000) studied the effect of pH on flavor release from a β-lactoglobulin solution, and they found that the release of methyl ketones, ethyl esters, limonene and myrcene was significantly slowed by changing pH to 6.0 and then to 9.0, but all the flavors were less retained when the pH value increased to 11.0. It was suggested that there was enhanced interactions between flavors and β-lactoglobulin at pH 3.0-9.0, as the flexibility of the protein molecules, the surface exposure of residues, and the unfolding of peripheric α-helix and β-sheet were likely to increase as pH increased from 3.0 to 9.0. However, at pH 11.0 the tertiary structure of the protein was largely modified due to alkaline denaturation (unfolding), and the flavor-protein interactions were weakened.

4.4 Other Factors

Other factors, including emulsion stability and emulsion types, were poorly addressed in the literature, but their effects on flavor release should not be neglected. For example, creaming in emulsions resulted in more oil phase transferred to the liquid-air surface, which then induced more flavor released to the headspace (Benjamin et al., 2012a). Studies on flavor release from emulsions of different types showed that the transfer rates of diacetyl toward the headspace was higher in an O/W emulsion than in a W/O emulsion stabilized with the same emulsifier (Salvador et al., 1994). The inclusion of a water phase in a fat blend could reduce the headspace

concentrations of butanoic acid and hexanoic acid (Shiota et al., 2011). Nevertheless, a perception test failed to identify any difference between O/W and W/O emulsions with same compositions (Bakker and Mela, 1996).

5. Effect of Mouth Conditions on Flavor Release and in vitro Studies

5. 1 Effect of Saliva on Flavor Release

In the oral cavity, the releasing behaviors of flavors from emulsions are mainly influenced by saliva. Apparently, dilution of food with saliva will first disturb the partition and mass transfer of flavors in aqueous phase and oil phase, leading to different releasing kinetics. Many studies conducted either *in vitro* or *in vivo* showed a significant decrease in flavor release with increase of saliva volume (Hansson, 2003; Mehinagic, 2004; van Ruth and Roozen, 2000), possibly because of the interaction between flavor compounds and the ingredients in the saliva, especially proteins (van Ruth et al., 2001). However, there are other flavor compounds which are released more to the headspace of the matrix-saliva mixture with a high proportion of saliva, probably due to the salting-out effect (Deibler et al., 2001). With the presence of many enzymes (e.g. α-amylase, lipase, esterase) in saliva, esters, thios and aldehydes were reported to experience enzymatic conversion to other flavors upon contact with saliva for some time (Buettner, 2002a,b), and losses of some flavors may also occur (Buettner, 2002b). Several studies reported that a full saliva containing salts, mucin, enzymes, etc. could enhance flavor release (Benjamin et

al., 2012a; Mao et al., 2013b). Moreover, the temperature change induced by salivation should also be taken into account when studying flavor release in mouth (Benjamin et al., 2012a). On the other hand, many studies have reported that emulsion properties can be greatly influenced by human saliva, which can then influence flavor release. Studies conducted on emulsion stabilized either by proteins (e.g. β-lactoglobulin, sodium caseinate, lactoferrin) or surfactants (e.g. tween 20, SDS) showed salivas can induce emulsion flocculation (bridge flocculation or depletion flocculation), and finally phase separation (Vingerhoeds et al., 2005; Sigh and Sarkar, 2011). The presence of different salts in human salivas can also modify the interfacial charge of droplets and the pH of the emulsions (Sakar et al., 2009). The effect of saliva on emulsion properties have been thoroughly discussed in the review of van Aken et al. (2007).

5.2 Model Mouths and Throats

Due to the availability of human samples, and the large variability of the samples among individuals, it is essential to develop mouth simulators (model mouth) to predict flavor release. The advantage of *in vitro* studies is that each oral parameter can be controlled, facilitating the understanding of its individual role on flavor release. Several model mouths have been developed in the past two decades, each of which can mimic the oral process to some extent. In this part of the review, we only introduce some model mouths/throat suitable for emulsion studies. Comparison of more mouth models can be found in the review of Salles et al. (2011).

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The simplest apparatus for studying flavor release in oral cavity was developed by van Ruth et al. (1994) (Figure 2A). The model mouth contained a sample flask (70 mL) which was surrounded by a water jacket with circulating water (37 °C), and sample-saliva mixture was added to the flask during experiments. A constant flow of nitrogen (at the flow rate of 20 mL/min) drove flavor release from the samples, and also flushed the headspace to a Tenax trap. The trapped flavors were later thermally desorbed and analyzed by GC. A more delicate model mouth was proposed by Elmore et al. (1996) (Figure 2B). The apparatus contained two taps on the top, which allowed the bottom part of the system to be removed without disturbing the gas flow to the MS detector. By turning on and off the taps, one can carefully monitor the start and finish time of the experiment. The apparatus consisted of a magnetic stirrer, which facilitated the mixing of liquid foods in the samples vessel (~125 mL) and simulated typical shear force in the mouth (~50 s^{-1}). Benjamin et al. (2012b) designed a novel model mouth to study the effect of tongue pressure on flavor release (Figure 2C). The artificial tongue was made of elastomer rubber, and it can be driven up and down vertically through the glass shaft (connected to an actuator), which simulated the tongue flattening against the hard palate during eating. Different patterns of tongue movement (ramp, pulse or sine wave) can be applied, and the tongue pressure was monitored by a sensitive pressure sensor attached to the bottom centre of the mouth chamber.

As discussed previously, a thin layer of liquid food can remain on the surface of pharynx, which

also contributes to flavor perception. Weel et al. (2004a) and Boelrijk et al. (2006) attempted to develop a model throat to study flavor release from throat *in vitro*. The main part of the model throat is a vertical glass tube (surrounded by a water jacket), with a 3mm thick tube of rubber (with clamp) in the middle which can control the liquid-saliva mixture to be flowed down through the tube (Figure 2D). When the samples flowed down and drained, a thin layer of the sample can form on the surface of the tube. Then air flow (1 L/min) enters the tube from the bottom and move upward, and the flavor released from the thin layer is flowed out with the air to the MS detector.

6. Novel Emulsions for Controlled Flavor Release

Due to their simple structures and compositions, conventional emulsions are prone to flocculation, coalescence, creaming and separation. It is thus difficult to achieve controlled or targeted release/delivery of active compounds incorporated in unstable emulsions. Therefore, novel emulsions with tailor-made structures in the water phase, oil phase and interface have been developed, and they can be used to deliver flavor compounds in a more controlled manner.

6.1 Multilayer Emulsion

A multilayer emulsion is an emulsion in which the droplets are surrounded by two or more layers (Figure 3), which is generally prepared through a layer-by-layer technique (LBL). The LBL technique normally consists of two (or more) steps of layer adsorption: a charged emulsifier is

firstly deposited onto the droplets surface; then an oppositely charged emulsifier or polymer is attracted by the previously adsorbed layer. As the formation of multilayer interface is mainly driven by electrostatic forces, ways that can modify these forces, such as pH adjustment, ionic strength modification, have been proved to alter the structures of the interfacial layer (e.g., layer thickness, compactness, charge density) (Guzey and McClements, 2006).

Emulsions containing oil droplets surrounded by a multilayered interface have been reported to exhibit better stability against pH change, heating, freeze-thawing, high ionic strength, etc. (Guzey and McClements, 2006), and they are more effective in protecting nutrients from degradation and improving nutrient adsorption (Djordjevic et al., 2007; Hou et al., 2011; Zhang and Zhong, 2015). More importantly, the multilayer emulsions can be designed to have different responsiveness to environmental stresses, and thus allow better controlled delivery of the active compounds incorporated (Guzey and McClements, 2006; Mao and Miao, 2015). Mao et al. (2013b) prepared WPI-pectin multilayer emulsions for the delivery of volatile flavor compounds, and they found that flavor was released at a lower intensity in the multilayer emulsion than that in a WPI single layer emulsion. When the pH of the emulsion was adjusted, the pectin could be detached from the interface, leading to higher release rates and enhanced intensity of the flavors. In a lecithin-pectin stabilized emulsion, the release of limonene was reduced by increasing the amount of pectin and lecithin added (Yang et al., 2011). When multilayer emulsions were

spray/freeze-dried, a greater flavor retention could be obtained (Kaasgaard and Keller, 2010; Gharsallaoui et al., 2012). Under oral conditions, single layer (β-lactoglobulin) emulsions presented fast release of flavor compounds upon the addition of small amount of saliva (due to emulsion destabilization), whereas the multilayer (β-lactoglobulin-pectin) emulsion was only slightly affected by saliva and presented almost the same flavor release profile as the primary emulsion without saliva (Benjamin et al., 2013).

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6.2 Gelled Emulsion

In a gelled emulsion, oil droplets are trapped within gel particles (Figure 3), which act as a barrier to slow the mass transfer and diffusion of the compounds incorporated. The preparation of gelled emulsions usually involves two steps: first, a conventional O/W emulsion is produced through homogenization; second, the emulsion is mixed with a biopolymer solution (biopolymer can be also present in the water phase of the emulsion), which gels by adjusting pH, salt concentration, temperature, adding enzymes or applying mechanical forces (McClements, 2010). Lipophilic functional ingredients can be dissolved in the oil droplets, and incorporated in the gel network. As the lipophilic ingredients are isolated from the external environment, they usually have improved chemical stability (e.g. oxidation). On the other hand, it is possible to get tailor-made structures (e.g., composition, dimensions, hardness, permeability) of the gel with different responsiveness to environmental stresses, and thus allow controlled-target release of the

incorporated ingredients.

Lee et al. (2006) prepared protein stabilized emulsion gels (induced by microbial transglutaminase) and found that flavor retention ranging from 60% to 100% could be obtained in emulsion gels, whereas only 5% to 25% was obtained in ungelled emulsions when stored at 37°C for 72 h. Mao et al. (2014) explored the effect of rheological properties of emulsion gels on flavor release. They reported that flavors released at lower rates and had lower air-gel partition coefficients in emulsion filled protein gels than those in their ungelled counterparts. In gels with a higher WPI content which were more elastic, and the incorporated flavors had reduced release rates and partition coefficients. An increase in the oil content resulted in more elastic but brittle gels, showing however a minor influence on the release rate of the flavors. Malone and Appelqvist (2003) designed biopolymer-gelled particles with encapsulated oil droplets containing flavors. They found that the initial maximum flavor release in the mouth was reduced due to the slower mass transfer of flavor molecules through the gel particles. The release behaviour can be modified by changing droplet size or oil phase volume of the emulsions. When the gel was broken down in a controlled manner by physiological triggers, such as mechanical force, melting, and enzyme treatment, a controlled release profile was obtained. For example, flavor compounds exhibited a faster release in a gelatinised amaranth amylose gelled emulsion than that in a gelatinised wheat starch gelled emulsion when the starch gel was broken down by

α-amylase. Flavor release from the gel was highly associated with the release of oil droplets during oral processing, and a change in droplet size allows modified flavor release. Studies on oral processing suggested that the release of oil droplets increased with droplet size (Guo et al., 2014), and smaller oil droplets were more firmly entrapped into the gel matrix, even under strong mechanical processing (Guo et al., 2013).

6.3 Multiple Emulsion

In a multiple emulsion, the droplets of the dispersed phase contain even smaller dispersed droplets (Figure 3), such as water-in-oil-in-water (W/O/W) emulsion and oil-in-water-in-oil (O/W/O) emulsion. A multiple emulsion has different layers deposited on both oil-water interface and water-oil interface. Multiple emulsions are normally prepared using a two-step homogenization method: the inner W/O (or O/W) emulsion is firstly produced, which is then dispersed into the outer dispersed water (or oil) phase (Garti, 1997; Garti and Bisperink, 1998).

A multiple emulsion contains two water (or oil) domains, which prolong the diffusion length of compounds incorporated in the inner dispersed phase, leading to reduced release rate and higher encapsulation efficiency of the core material. Moreover, it becomes possible to effectively control the release of both hydrophilic and lipophilic ingredients in a single system. W/O/W emulsion can be used in fat-reduced system, as the oil content of the oil phase can be partially displaced by the inner water phase. Dickinson et al. (1994) made a protein-stabilized W/O/W

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emulsion and found that the release rate of butanol was reduced by a factor of two, in comparison to the release rate of the volatile from an equivalent aqueous solution of butanol. Similar to conventional emulsion, flavor release from multiple emulsion is also affected by the emulsifier used. In an O/W/O emulsion with gum arabic as the inner droplets stabilizer, a combination of PGPR and Span 80 on the outer interface can reduce flavor release to a greater extent than PGPR alone (Cho and Park, 2003). When a multiple emulsion was spray-dried, the preparation can be stored for longer time with less flavor loss (Brückner et al., 2007).

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6.4 Pickering Emulsion

Pickering emulsions are defined as emulsions, either O/W or W/O type, stabilized by solid particles at the interface (Aveyard et al., 2003). Generally, they have similar physicochemical properties as conventional emulsions, and they can be used to substitute conventional emulsions in most cases, particularly in the case where a surfactant-free system is essential. The interface of a Pickering emulsion is mechanically stronger, and they can provide sufficient steric repulsion force to inhibit droplet coalescence. Different types of silica are widely used to produce Pickering emulsions, and in the food industry, cellulose nanocrystals (Kalashnikova et al., 2011), chitin nanocrystals (Tzoumaki et al., 2011), starch particles (Yusoff and Murray, 2011), and flavonoid particles (Luo et al., 2011) have proved to be suitable Pickering stabilizers.

With good physical stability, Pickering emulsions have been reported to better deliver salts

(Frasch-Melnik et al., 2010), curcumin (Wang et al., 2014), fatty acids (Ruiz-Rodriguez et al., 2014), retinol (Ghouchi Eskandar et al., 2009).Wang et al. (2012) made $CaCO_3$ particle stabilized emulsions, and they tested the release behaviour of limonene. The authors reported that non-encapsulated limonene released very fast and most of the flavor released in less than 5 min, while limonene in the Pickering emulsion exhibited slower release with a retention time over 40 min. It was possible to break up the emulsion by dissolving $CaCO_3$ particles in the acidic solution, and thus triggered faster release of limonene. The Pickering emulsions using biopolymer-based particles as stabilizers need to be further studied as potential vehicles for flavour delivery and controlled release due to their exceptional stability properties.

6.5 Self-assembly Structured Emulsion

Many emulsifiers, especially the small molecular surfactants, can not only form the interfacial film on the droplets, but also organize themselves into micelles (reverse micelles), hexagonal, lamellar, and other self-assembled structures (Krog and Sparsø, 2004), which then structure the emulsions (Figure 3).

Emulsifier micelles or reverse micelles can largely change partition coefficients of flavor compounds, as they are capable of solubilising them in their hydrophobic (hydrophilic) domains (Rabe et al., 2003a). van Ruth et al. (2002a) have studied the effect of tween 20 on flavor release, and they found that tween 20 micelles can retain hydrophobic volatiles, and tween 20 reverse

micelles can retain hydrophilic volatiles. In another study, the formation of tween 80 micelles and PGPR (polyglycerol polyricinoleate) reverse micelles was shown to protect citral flavor from degradation (Choi et al., 2010).

Monoglycerides (MGs) are oil-soluble emulsifiers and they can self-assemble into lamellar, cubic or hexagonal phases in contact with water or oil. Recent studies revealed that the self-assembled structures can provide protection for sensitive ingredients (Sagalowicz et al., 2006; Larsson, 2003). The application of MG self-assembled structure to control flavor release has been reported recently. For MG structured W/O microemulsions, Landy et al. (2007) reported that lipophilic volatile compounds are retained at a greater extent in the MG structured emulsions, compared with the unstructured W/O emulsions. In MG structured oil-in-water gel systems, Calligaris et al. (2010) discovered that the equilibrium concentration of limonene in the headspace of MG gel was significantly lower than that of a conventional emulsion. Phan et al. (2008) made MG structured O/W emulsions with low oil content, in which delayed volatile release was also observed. Mao et al. (2013a) found that flavors had lower headspace concentrations and partition coefficients in MG structured emulsions than in conventional unstructured emulsions, and that tween 20 stabilized emulsions (with MG) were more capable to slow flavor release than WPI stabilized emulsions (with MG) (Mao et al., 2012). However, in an oil-reduced system the role of monoglyceride crystalline structure is weakened and lipophilic

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flavors are released at higher rates and of higher intensity (Mao et al., 2013a).

7. Conclusion

Extensive studies have been performed to understand flavor release from food emulsions and thereby to design effective colloidal dispersion systems with improved physical-chemical stability and desired flavour kinetic release profiles. Flavor release from emulsions is affected by various factors, including ingredient composition and emulsion structures; these factors can function either individually or synergistically. It is now quite clear that oil plays a dominant role on flavor release, while emulsifiers and other ingredients are all likely to interact with flavor compounds and lead to altered flavor release. Modification of emulsion structures has been regarded as a novel way to modulate flavor release, while maintaining the composition of the food emulsion to a large extent. Structured emulsions, those which have been proved as effective delivery systems for many functional food ingredients, could be also working well to deliver flavor compounds. Therefore, emulsions have a great potential for applications in functional foods (e.g., low fat, low salt, low sugar products) with favorable flavor characteristics. However, due to the complexity of food matrices, the diversity of flavor compounds characteristics, and the miscellaneous responses of emulsion structures during processing, transportation and consumption, our understanding about flavor release from food emulsions is rather limited, and the overall process is still very difficult to control. Moreover, information obtained from studies on model systems does not fully reflect flavor release behaviour from a real food matrix. When

flavor perception is concerned, more well-designed models are required to understand the effect of oral conditions on emulsion structures and flavor release, and to fully unravel the relationships between texture and flavor perception.

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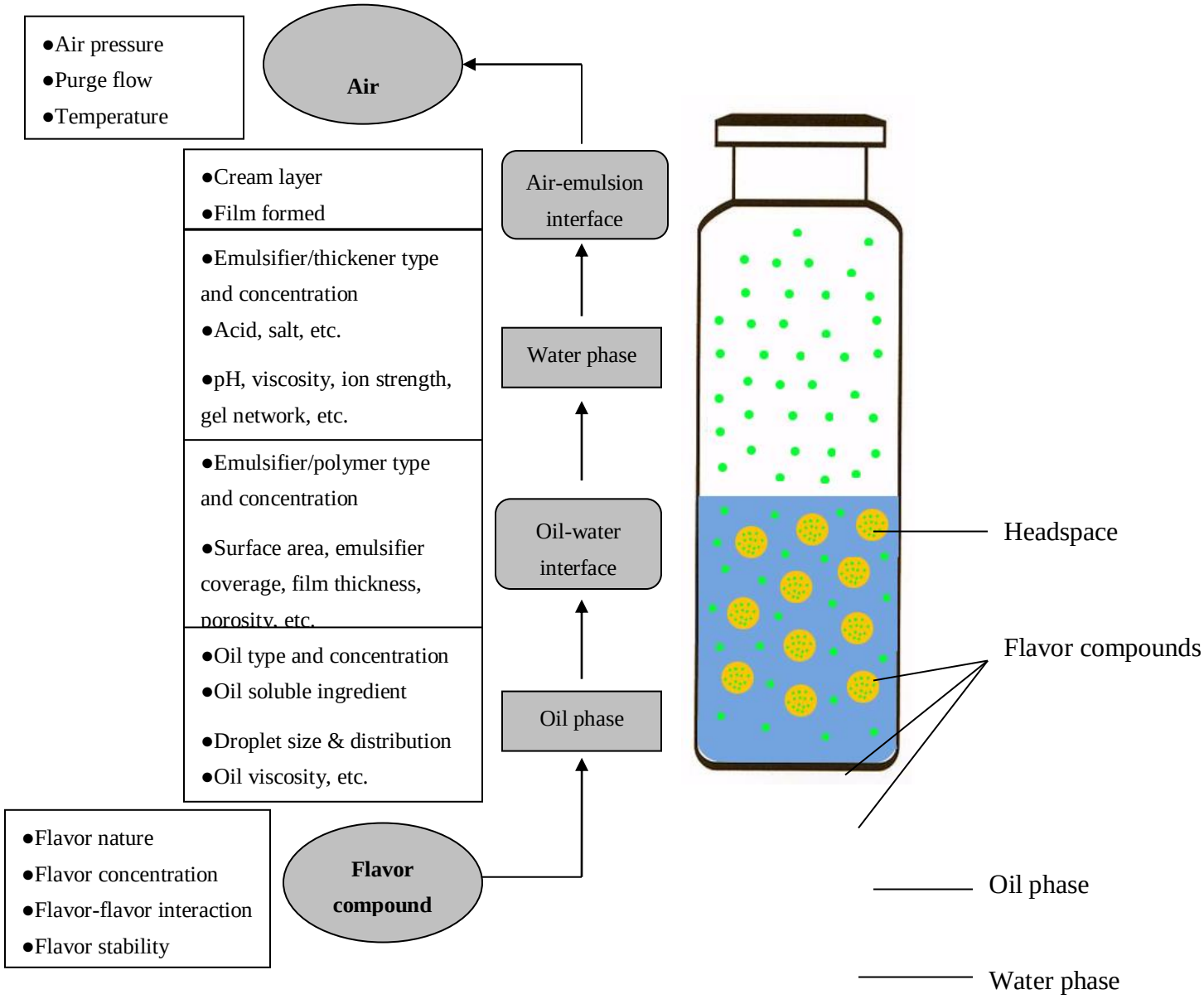
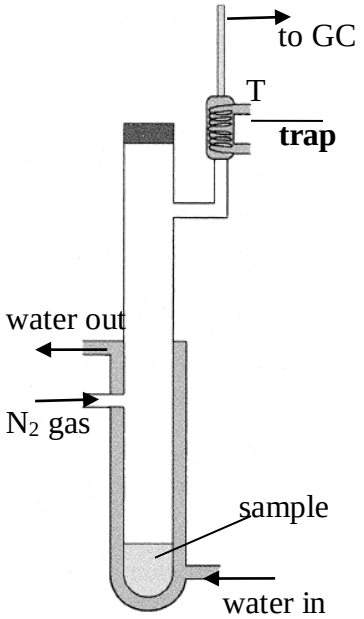


Figure 1. Schematic diagram of flavor release from O/W emulsion and affecting factors.



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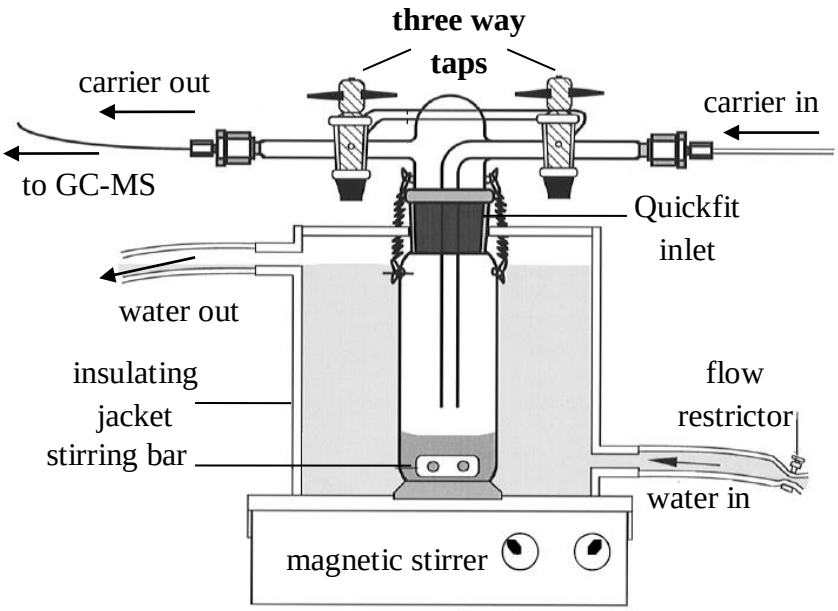

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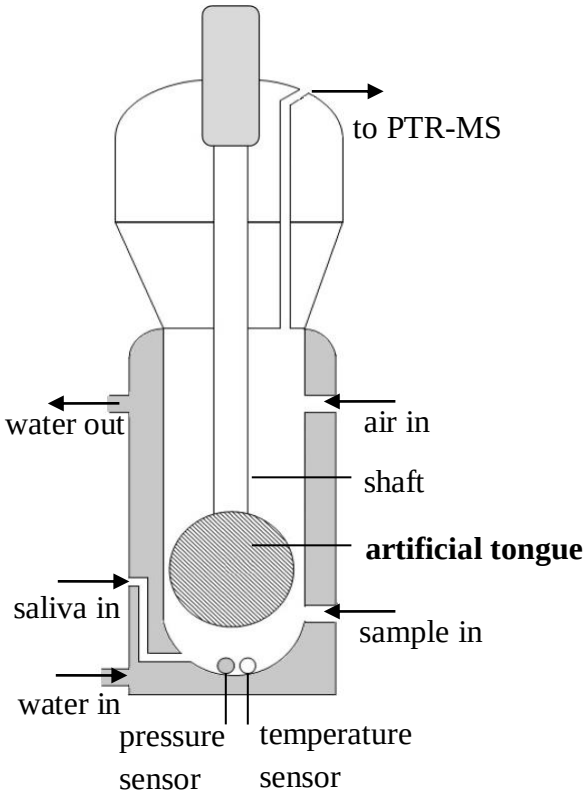
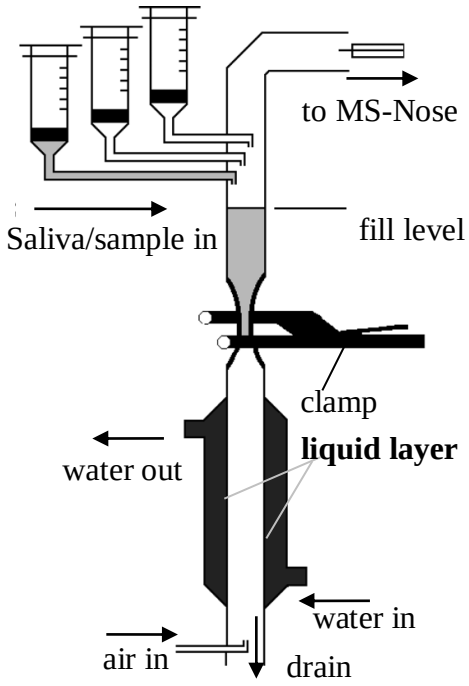


C



D

Figure 2. Representative model mouths (A, B, C) and throat (D) for *in vitro* studies of flavor release from emulsions. A- van Ruth, et al. (1994); B- Elmore, et al. (1996); C- Benjamin, et al. (2012b); D- Weel, et al. (2004a)

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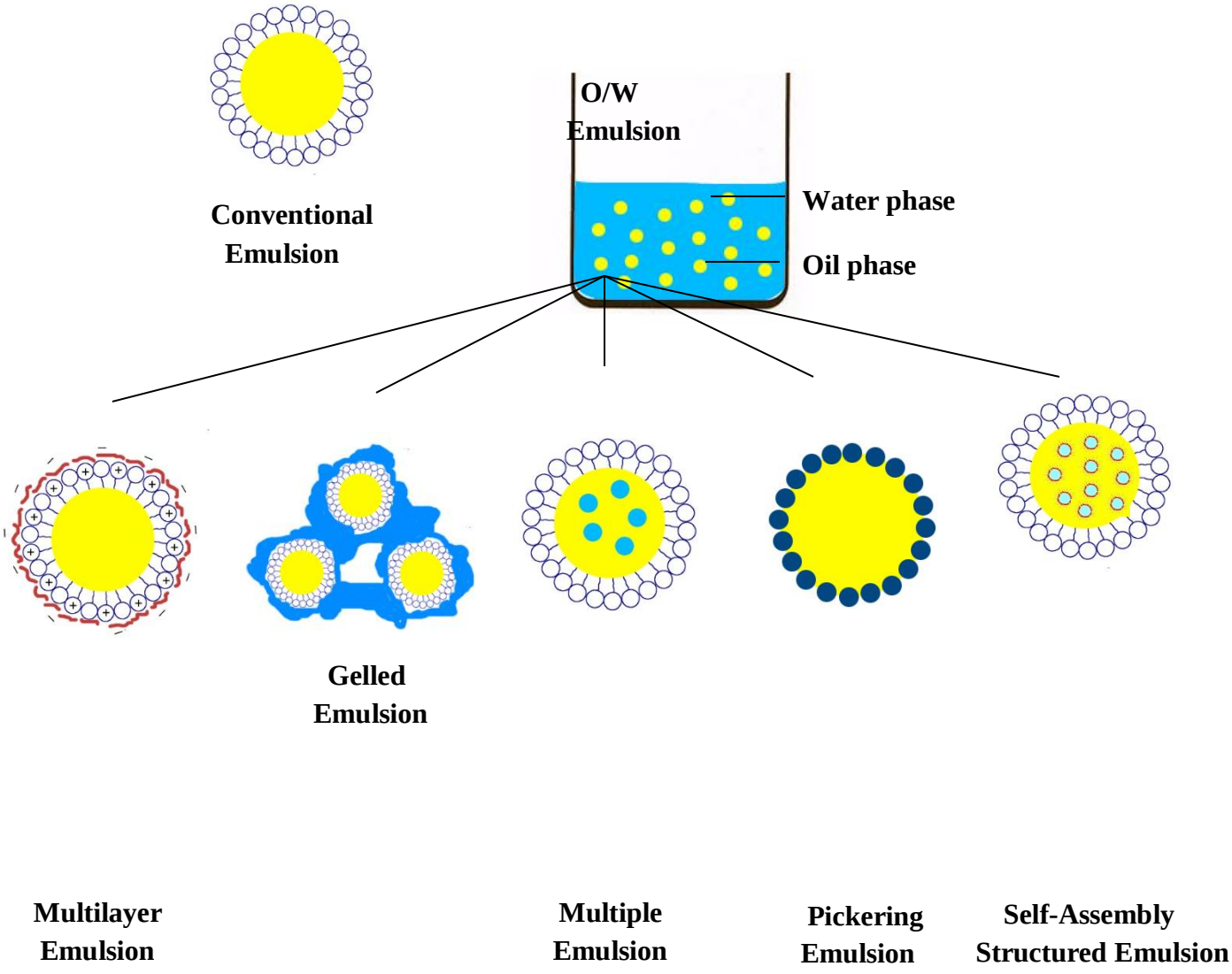

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Figure 3. Different types of emulsions suitable for flavor delivery. In the self-assembly structured emulsion, only the emulsion containing reverse micelles is illustrated.