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Encapsulation Efficiency of Food Flavours and Oils during Spray Drying

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Microencapsulation is a rapidly expanding technology which is a unique way to package materials in the form of micro- and nano-particles, and has been well developed and accepted within the pharmaceutical, chemical, food and many other industries. Spray drying is the most commonly used encapsulation technique for food products. A successful spray drying encapsulation relies on achieving high retention of the core materials especially volatiles and minimum amounts of the surface oil on the powder particles for both volatiles and non-volatiles during the process and storage. The properties of wall and core materials and the prepared emulsion along with the drying process conditions will influence the efficiency and retention of core compounds. This review highlights the new developments in spray drying microencapsulation of food oils and flavours with an emphasis on the encapsulation efficiency during the process and different factors which can affect the efficiency of spray drying encapsulation.

Keywords Encapsulation efficiency; Microencapsulation; Retention; Surface oil content; Volatiles; Wall materials

INTRODUCTION

Following the first commercial use of microencapsulation in 1954 to create a carbonless copy paper,[1,2] different encapsulation techniques were developed and accepted within the pharmaceutical, chemical, cosmetic, and food industries.[3,4] Microencapsulation is the process by which active ingredients (core materials) such as food oils and flavours are packaged within a secondary (wall) material.[5–7] The size of particles formed through encapsulation may be classified as:[8] macro (>5000 μm): micro (1.0–5000 μm); and nano (<1.0 μm). Capsules below 1.0 μm in size are frequently referred to as nanocapsules, which are often made by very specialized nanoencapsulation methods.[9–14]

Two main structures are single-core and multiple-core microcapsules (Figure 1). The former one is typically produced by complex coacervation, fluidized bed drying, droplet co-extrusion, and molecular inclusion, and has high core loading (e.g., 90% of total capsule weight).[15–17] In multiple-core capsules, which are produced principally by spray drying, the core material is dispersed throughout the wall material and the central area is occupied by the void resulting from expansion of particles during the later drying stages,[18–21] as shown in Figure 1. Microcapsules with this structure often have a core loading of 20–30% of total capsule weight.[16] Several techniques including scanning electron microscopy (SEM) can be used to investigate the external and internal structures of microcapsules.[21–25]

There are many works in the literature dealing with general issues of microencapsulation, which are outside the scope of this discussion. For instance, authors such as McKernan,[26,27] Arshady,[5] Shahidi and Han,[2] Dezarn,[28] Gibbs et al.,[3] Augustin et al.,[6] Gouin,[29] and Desai and Park[7] have published some review papers related to the microencapsulation of food ingredients and recently, Madene et al.,[4] and Vega and Roos[30] have presented good information on encapsulated flavours and dairy ingredients, respectively. An overview of the microencapsulation process is presented in Figure 2 including different core and wall materials, encapsulation techniques and various aims of producing encapsulated food ingredients.

MICROENCAPSULATION OF FOOD FLAVORS AND OILS

The initial step in encapsulating a food ingredient is the selection of a suitable wall material, basically a film-forming biopolymer, from a wide variety of natural or synthetic polymers, depending on the core material and the characteristics desired in the final microcapsules.[8,31,32] For flavour and oil encapsulation in particular, the ideal wall material should have emulsifying properties; be a good film

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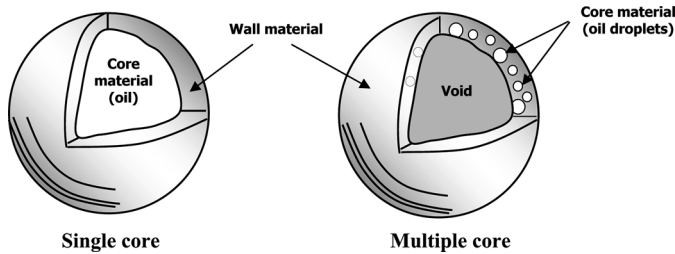
FIG. 1. Two different types of microcapsule structures.

former; have low viscosity at high solids levels; exhibit low hygroscopicity; release the flavour when reconstituted in a finished food product; be low in cost; bland in taste; stable in supply; and afford good protection to the encapsulated flavour and oil.[7,33–36] Because almost no wall material can meet all the properties listed, in practice they are used in combination with each other. Some types of wall materials along with their needed properties are presented in Figure 2. The commonly used wall materials in spray
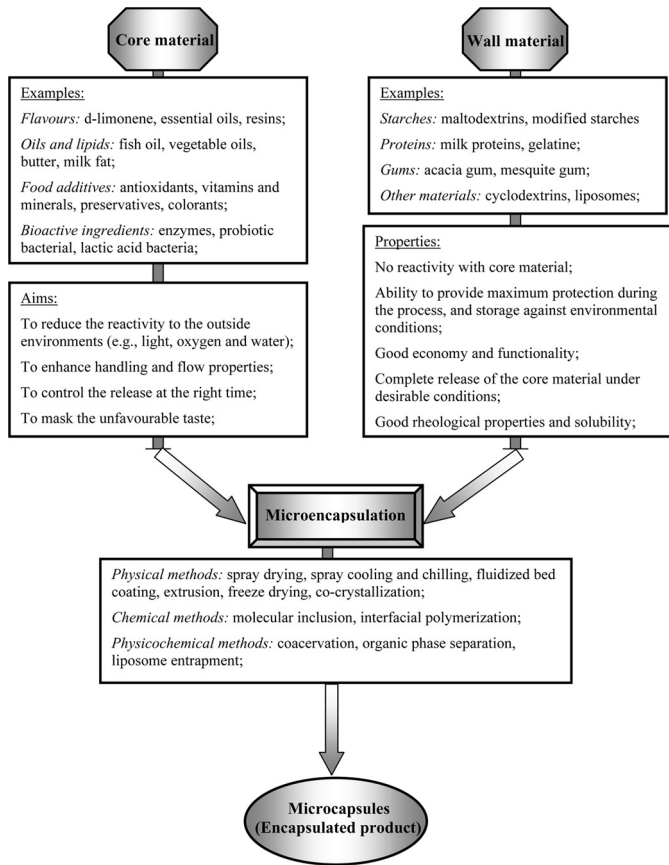

FIG. 2. Schematic description of the microencapsulation of food ingredients along with some example of core and wall materials, wall material properties, aims, and different techniques of the microencapsulation process.

drying microencapsulation of flavour and oils will be discussed later in section 4.1.

Microencapsulation can potentially offer numerous benefits to the food ingredients being encapsulated. Various properties of active materials may be changed by encapsulation. For instance, handling and flow properties can be improved by converting a liquid to a powdered encapsulated form. Hygroscopic materials can be protected from moisture and stability of ingredients that are volatile or sensitive to heat, light or oxidation can be maintained. Materials that are otherwise incompatible can be mixed and used safely together.[6,8,32,35,36] There are many different types of microcapsules being used as food additives such as encapsulated food flavours and edible oils. Some examples are given in Figure 2.

A vast majority of the flavour compounds used in the food industry are mainly in the form of liquid at room temperature. Also there is a need to incorporate some edible oils such as fish oil and many other vegetable oils into food products to increase the nutritional value of these products.[37,38] Most of these food oils exhibit considerable sensitivity to air, light, irradiation and elevated temperature.[39–41] Conversion of liquid flavours and edible oils to dry powders is an important application of microencapsulation in the food industry.[4,34,35] Also, one of the key aims for the microencapsulation of food oils and flavours is to control the release of these active ingredients until the right time.[4,42] Microencapsulated oils provide the convenience of a solid powder, with reduced volatility and less oxidation, and can be used in many different finished products such as cakes, beverages, etc.[2,35,43] Examples of commonly used encapsulated flavours and oils are citrus oils, artificial or natural flavours, essential oils and spices, tuna oil, fatty acids, soy oil, and sunflower oil.[44–48]

MICROENCAPSULATION BY SPRAY DRYING

Numerous techniques have been developed for the manufacture of encapsulated food ingredients, as some of them were given in Figure 2. Spray drying is the most commonly used encapsulation technique in the food industry,[19,20,32] and one of the oldest encapsulation methods, being used in the 1930s to prepare the first encapsulated flavours using gum Arabic as the wall material.[2] Also, spray drying and extrusion are the most popular processes for the microencapsulation of food flavours and oils.[49–51] The process of spray drying is economical and flexible, uses equipment that is readily available, and produces powder particles of good quality. Authors such as Re,[32] Sharma and Tiwari,[52] Reineccius,[19,20,53] and Bhandari[51] have published some good reviews on spray drying microencapsulation.

Carbohydrates, milk proteins, and new emerging biopolymers make up the three main classes of wall materials generally available and suitable for spray drying microencapsulation[34,54–56] that will be reviewed in section 4.1.

After selecting the suitable wall material, it must be rehydrated (sometimes with heating) in water.[57] This is particularly important for surface-active biopolymers to exhibit their emulsifying capabilities during emulsion formation.[58] It is desirable to use a pre-determined infeed solids level that is optimum for each wall material composition. When the wall material has been hydrated, the core material must be added to make a coarse emulsion, usually via high-speed mixing or high-shear emulsification by colloid mills. A 20–25% flavour load based on total solids of the wall solution is traditional in spray drying microencapsulation.[32,59–61] Then, final emulsion will be prepared by other emulsification methods including high-pressure homogenization, e.g., microfluidization.[62]

Following the preparation of the infeed emulsion, it will be pumped to the drying chamber of the spray drier. Two types of atomizers are widely used:[63,64] the high-pressure nozzle; and the centrifugal wheel. The industry is nearly equally divided between the use of these two types of atomizers.[20] Although each type of atomization has its own advantages and disadvantages, there is no literature suggesting that one type results in a better effect than the other.[2,64] As the atomized droplets fall through the hot air medium inside the drying chamber, they assume a spherical shape. For the spray drying encapsulation of food flavours and oils generally, co-current air flow is applied. The rapid evaporation of water from these droplets during surface film solidification keeps the core temperature below 100°C in spite of the high temperatures (>150°C) used in the process. The particles' exposure to heat is in the range of a few seconds at most.[19,20,26,52,65] Because core materials such as flavours, may contain many various components with different boiling points, it is possible to lose certain low boiling point aromatics during the drying process.[66–68] Spray-dried encapsulated powders typically have a very small particle size (generally less than 10 μm) with a multiple-core structure (Figure 1).

DIFFERENT PARAMETERS AFFECTING THE ENCAPSULATION EFFICIENCY DURING SPRAY DRYING

Successful encapsulation of flavours and oils should result in an encapsulated powder with minimum surface oil content on the powder particles and maximum retention of the core material, particularly volatiles, inside the particles. A need to optimise the retention of flavours and the encapsulation efficiency of fish oil and many other edible oils during spray drying motivated the considerable research that has been carried out over the last couple of years.[30,69–74] It is surprising how volatile flavour compounds are retained during spray drying without being lost to a large extent. The major constituent in the infeed emulsion is water, which evaporates during drying ($>90\%$), but yet the relatively more volatile flavour constituents are

nearly completely retained when optimum drying conditions are followed.[20,53,75,76] Two theories have been developed in this regard: According to the "selective diffusion" theory, when surface moisture of atomized droplet decreases to about 7 to 23 percent ($a^w < 0.90$), this dry surface acts as a semi-permeable membrane permitting the continued loss (or diffusion) of water but efficiently retaining flavour molecules.[19,20] As drying continues, the diffusivity of the flavours reduces dramatically compared with water molecules. Therefore, more losses occur during the earlier stages of spray drying. Here, the "relative volatility" theory is applicable: flavours with higher relative volatility than water will be lost more than those with lower volatility during the initial drying stage.[51]

Much of the information of volatile losses during the drying comes from studies on single droplets.[20,77–80] King and Hecht[81–83] have defined three regions where volatile losses occur: **(1)** during atomization, as there is a large surface area, turbulence and flowing/mixing within the sheets of the atomized emulsion; **(2)** after droplet formation when there is rapid water loss from the droplet, and its surface has not formed a stable selective membrane. Thus, volatiles diffuse with water to the droplet surface, particularly those with higher relative volatilities, and are lost to the drying air; **(3)** the final loss is when the water in the droplet exceeds its boiling point and bubbles formed inside the droplet most bursting out to the surface, taking volatiles with them. It is shown that losses during this third stage (i.e. during morphological development) are greater than during atomization and the beginning of surface drying (selective diffusion).
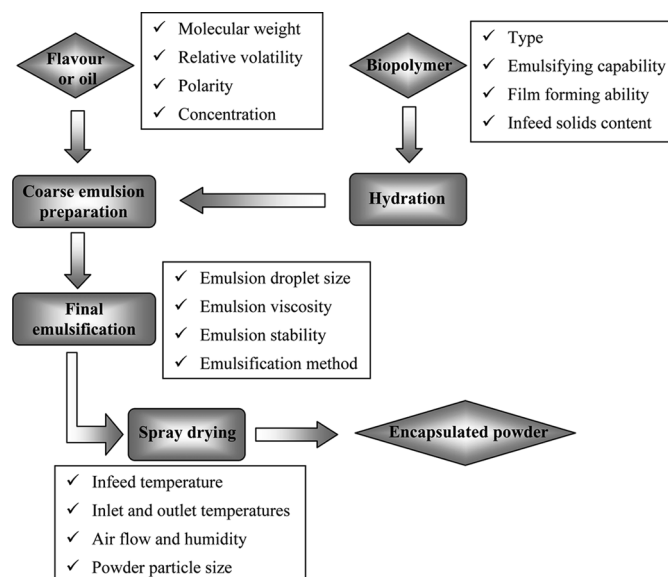

FIG. 3.   Flow diagram of spray-drying microencapsulation of food flavors and oils including the factors affecting the encapsulation efficiency.

In optimizing the process, there are at least four group of criteria that can be considered: **(a)** properties of the wall materials; **(b)** characteristics of the core materials; **(c)** specifications of the infeed emulsion; and **(d)** conditions of the spray drying. In Figure 3, a flow diagram of spray drying microencapsulation is presented with factors affecting the encapsulation efficiency at each step.

Properties of the Wall Materials

There are numerous wall materials available for use as flavour and oil encapsulating agents. For spray drying microencapsulation, in particular, the choice of wall material is critical as it will influence emulsion properties before drying, retention of the volatiles during the process and shelf-life of the encapsulated powder after drying. Among the available ingredients, the major wall materials used for spray drying applications are *carbohydrates* including modified and hydrolysed starches, cellulose derivatives, gums, and cyclodextrins; *proteins* including whey proteins, caseinates, and gelatine; and new *emerging biopolymers* such as products of Maillard reaction. A brief summary of these wall materials along with their properties and applications and related references is presented in Table 1.

Carbohydrates

Hydrolysed starches are depolymerised ingredients produced by hydrolysing starch with acid and/or enzymes.[84,85] These wall materials offer the advantage of being relatively inexpensive; bland in flavour; low viscosity at high solids; and excellent protection to encapsulated core materials such as orange oil, milk fat, soy oil and fish oil. The degree of protection is directly related to the dextrose equivalent (DE) of the hydrolysed starch, higher-DE systems are less permeable to oxygen and result in powders with higher encapsulation efficiencies.[25,32,47,54,86–89] These ingredients, however, lack any emulsifying properties and typically result in poor retention of flavours during spray drying.[33,90–93] For instance, Re and Liu[70] reported just 67% retention of Allylguaiacol by using maltodextrin DE 10 as the wall material compared with 94% retention by modified starch. Even substantial differences in flavour retention and shelf-life are observed for the produced hydrolysed starches from different sources.[94] Therefore, it is desirable to use them in combination with a surface-active biopolymer, such as esterified modified starches,[59,60,72,95,96] gum Arabic,[69,80,97–101] or milk proteins.[40,47,89,102] Blends of commercial wall materials have been evaluated by these workers, with the aim to obtain an effective spray drying encapsulation with high retention or encapsulation efficiency and low costs.

In order to give some emulsifying capabilities to starch molecules, side chains of lipophilic succinic acid are inserted into starch to produce modified starches.[103,104] Various forms of modified starches are used for flavour

and oil encapsulation such as Capsul, N-lok, Hi-cap and Encapsul.[105] Some workers have shown that wall materials based on modified starch leads to very good retention of volatiles and low amounts of unencapsulated oil at the surface of powder particles.[106–108] For example, Jeon et al.[109] investigated the encapsulation potential of native corn and barley starches (regular and waxy) and their chemically modified counterparts (Succinylated and Octenyl Succinylated starches) to minimize the evaporative flavour loss and to improve flavour stability. They found that chemically modified (Succinylated) corn and barley starches are more effective than the native starches in the flavour retention. In particular, Succinylated regular starches showed better retention ability than waxy starches. Also, modified starches can be used at high infeed solids level (compared to gum Arabic) and may afford outstanding emulsion stability.[103]

Among wall materials, Gum acacia (Arabic) has been the most popular and common ingredient for spray drying encapsulation of oils and flavours, since it has emulsifying properties and provides excellent volatiles retention during the drying process.[65,98,110–115] But in recent years, its high cost, limited availability, and the impurities associated with it have been deterrents to the use of gum Arabic despite its exceptional capabilities, and researchers have tried to use a blend of gum Arabic (GA) with other wall materials and/or to replace GA completely. For example, a combination of gum Arabic and maltodextrin (MD) was reported to be effective for the encapsulation of cardamom oil,[97] citral and linalyl acetate,[69] citrus oils,[98] soy oil,[116] rice flavour,[68] fatty acids,[21,117] pine flavour,[56] and bixin.[118] These workers have shown that maltodextrins can successfully replace a part of GA as wall material and they have determined the best ratios of MD:GA. Beristain and his co-workers[119–123] evaluated the performance of mesquite gum as compared to gum Arabic in the spray drying microencapsulation of orange peel oil and cardamom essential oil. Their results showed that a blend of 60% gum arabic and 40% mesquite gum encapsulated 93.5% of orange peel oil,[120] and a mixture of 40% mesquite gum and 60% maltodextrin was able to encapsulate 84.6% of the oil.[121] This confirms the good emulsifying properties and encapsulation ability that qualifies mesquite gum as an alternative wall material.

Cyclodextrins have also been used in spray drying encapsulation of food oils and flavours. They are cyclic molecules containing six (alpha-), seven (beta-) or eight (gamma-) glucose monomers that are produced from starch.[50,124] These monomers are connected to each other, giving a ring structure that is relatively rigid and has a hollow cavity with the ability to encapsulate other molecules. Many reports have demonstrated that inclusion complexes are virtually completely stable to oxidation compared to other wall materials.[61,71,93,125] In a study by Reineccius

TABLE 1

Different wall materials used in spray-drying microencapsulation of food oils and flavors

Wall materials		Properties	Examples	Encapsulated flavors and oils	Cited references
Carbohydrates	Hydrolyzed starches	Very good oxygen-barrier, cheap, low viscosity at high solids; no/limited emulsion stabilization	Corn syrup solids, maltodextrins	Citral and linalyl acetate; ethyl caprylate; cheese aroma; linoleic acid; orange peel oil; lemon oil	[54, 67, 69, 84, 86, 87, 90–92, 96, 134, 164, 171, 181, 182]
	Modified starches	Very good emulsion stabilization, inexpensive; sometimes varying quality, not universally usable owing to regulatory situation	Capsul, N-lok, Hi-Cap	Meat flavor; fish oil; orange oil; d-limonene; l-menthol; butter oil; cream; black pepper oleoresin; vitamin E	[44, 59, 60, 72, 95, 106–109, 112, 116, 135, 158, 180, 183]
	Gums	Good emulsions, very good retention of volatiles; varying quality, price depends on availability, sometimes impurities	Arabic gum, mesquite gum	Essential oils; monoterpens; orange peel oil; cardamom oil; vegetable oils; cardamom oleoresin; linoleic acid; bixin; short-chain fatty acids; lipids; acetyl pyrroline; soy oil; d-limonene; ethyl butyrate	[21, 68, 74, 80, 99, 100, 112–116, 118–123, 131, 174]
	Cyclo-dextrins	Very good inclusion of volatiles, excellent oxygen barrier; relatively expensive	α-, β-, α-Cyclodextrins	Pine flavor; shiitake flavor; d-limonene; ethyl hexanoate; caraway fruit oil; lemon oil	[56, 61, 71, 125–128, 186]

Proteins	Milk proteins	Very good emulsions; expensive, being dependent on other factors such as pH and ionic strength, allergenic potential	Whey proteins	Milk fat; linoleic acid; soy oil; ethyl butyrate; ethyl caprylate	[24, 46, 67, 88, 129, 141, 142, 146, 150, 151]
			Caseinates	Fish oil; soy oil	[40, 46, 47, 89, 140, 153]
			Skim milk powders	Oregano and marjoram flavors; caraway essential oil	[48, 102]
Other biopolymers	Other proteins; Soluble soy polysaccharides, chitosan, alginates, Maillard products, modified celluloses, gelatine		Soy proteins	Wheat germ oil; evening primrose oil; PUFAs; fish oil	Orange oil [143, 144]; [38, 73, 132, 133, 138, 139]

et al.,[126] they found that γ-cyclodextrin generally functioned better than α- and β-cyclodextrins in terms of initial flavor retention. On storage, however, their results showed that losses of volatiles were greatest for γ-cyclodextrin and least in the case of α-cyclodextrin. Bhandari and his co-workers have also published some works in this field by using lemon oil[127,128] and d-limonene[129] as the core material and beta-cyclodextrin as the wall material. Their results showed that the retention of lemon oil reached a maximum at the lemon oil to beta-cyclodextrin ratio of 6:94; however, the maximum inclusion capacity of β-cyclodextrin and a maximum powder recovery were achieved at the ratio of 12:88, in which the β-cyclodextrin complex contained 9.68% (w/w) lemon oil.

There have been some new studies on the use of carbohydrates in encapsulation. For instance, Zeller et al.[130] have described an alternative encapsulation technique based on physical adsorption of flavours onto the surface of highly porous carbohydrates. Also, Perez-Alonso et al.[131] have worked on the estimation of the activation energy of carbohydrate polymers blends as selection criteria for their use as wall materials. They showed that a mixture of 66% gum Arabic + 17% mesquite gum + 7% maltodextrin had the highest activation energy, so the best protection of encapsulated powders against oxidation. To this end, some workers have tried to apply novel biopolymers in spray drying encapsulation of food flavours and oils such as alginates,[132] chitosan,[133] soluble soy polysaccharides,[60,134,135] sucrose and flour,[136,137] products of Maillard reaction,[138] and modified cellulose.[73,139] This studies open new areas of research and need more works to be done.

Proteins

Functional properties of proteins including solubility, film formation, the ability to interact with water, emulsification and stabilization of emulsion droplets, exhibit many of the desirable characteristics for a wall material.[102] One of the commonly used proteins is gelatine.[2,140] In recent years, however, other proteins, particularly soy proteins, and milk proteins such as whey protein concentrate (WPC), skimmed milk powder (SMP), and caseinates have also been explored in many studies for their potential as new wall materials for spray drying encapsulation of flavours and oils. These proteins change their structure during emulsification through unfolding and adsorption at the oil-water interface and by forming resistant multilayer around oil droplets and also with the help of repulsive forces, make significantly stable emulsions which are critical for encapsulation purposes. Investigations have proven proteins to function well for encapsulating anhydrous milk fat,[24,141,142] orange oil,[143,144] soy bean oil,[46,89,145] caraway essential oil,[102] fish oil and fatty acids,[40,47,146] and oregano and marjoram flavours.[48]

For instance, Kim et al.[143,144] found that soy protein isolate (SPI) was the most effective wall material for retaining orange oil during spray drying (effectiveness = 85.7%), followed by sodium caseinate (81.7%), gum Arabic (75.9%), and whey protein isolate (72.2%). In another work, Bylaite et al.[102] showed that encapsulation efficiency was higher in WPC-based wall materials compared to SMP. Recently, Jimenez et al.[146] by encapsulating linoleic acid through spray drying with whey proteins found that encapsulation efficiency was about 89.6% with a surface oil content of 1.77 g/100 g powder and the produced microcapsules were stable over 60 days at aw = 0.743–0.898. It should be noted that the main problem with using proteins as wall materials for food flavours and oils is their performance being dependent on some other factors such as pH, ionic strength, and temperature.[147,148] For example, if the pH of initial emulsion prepared for spray drying encapsulation reaches the iso-electric point of the used protein, the biopolymer will lose its surface-active properties and the resultant emulsions becomes unstable, not suitable for encapsulation. Other biopolymers such as modified starches and modified celluloses are advantageous in this regard since their functionality is less affected by these factors.

Researches are also investigating the combination of proteins with different carbohydrates as wall materials. For example, it has been shown that a blend of whey proteins with maltodextrin and corn syrup solids[25,54,102,142] and lactose,[67,149–152] soy protein with maltodextrin,[18,70] sodium caseinate with lactose[153] and carbohydrates blends,[47,88,89] and WPI or SMP with maltodextrin[102] increases the retention of volatiles and the effectiveness of food oil encapsulation during spray drying process. An important issue in using biopolymers as emulsifiers is that due to slow adsorption kinetics, they can not produce very fine emulsions (real submicron emulsions), which are fundamentally favourable in spray drying encapsulation.[62] The other important factor is the total solids content of the emulsion that will be discussed along with other roles of the emulsion on encapsulation efficiency during spray drying in section 4.3.

Properties of the Core Materials

Retention of Volatiles

The loss of some volatiles including flavours during spray drying encapsulation is inevitable. Other than properties of the used wall material, some features of the core material will also affect the retention during the process. The fact that both "molecular weight" and "vapour pressure" of the flavour compounds have an influence on their retention during spray drying is both obvious and well documented in the literature.[19,20,32,76,154]

Molecular weight. is a reasonable representation of molecular size, which actually is the primary factor determining diffusion.[154,155] The increase of molecular size generally results in slower diffusion rate, subsequently, the molecules will take more time to reach the atomised droplet surface during drying, particularly initial stages, and retention will increase. A second factor promoting the retention of large favour molecules is that the surface of the droplet becomes impermeable to them more quickly during drying, when diffusions effectively stops at low moisture content. Both of these factors favour the retention of larger molecular weight (size) volatiles. This behaviour has been observed for spray drying of two different esters with gum Arabic:[65] ethyl hexonate (MW = 144) was always better retained than ethyl butyrate (MW = 116). The same trend has been noticed by Voilley et al.[156] for a mixture of 16 aroma compounds encapsulated in glucose, maltose or corn syrup solids. The retention rate of isoamyl butyrate (MW = 158) was higher than that of ethyl butyrate (MW = 116) or ethyl propionate (MW = 102) in all tested wall materials, except in maltose and corn syrup solid with DE 28.5.

Relative volatility. plays a secondary role in determining flavour retention owing to its influence in controlling flavour loss until the droplet surface becomes semi-permeable. Volatility reflects the ability of a compound to reach the gaseous phase and can be evaluated by measuring the vapour pressure of the pure compound.[19,154] Relative volatility of a compound is calculated with respect to water.[51] Bangs and Reineccius[90] have shown that retention of octanol, octenol, octanon and octanal were related to their relative volatility when they were encapsulated with maltodextrin and spray dried. Retention of these four different aroma compounds was reported based on their relative volatility in the mixture before drying: the higher the relative volatility, the lower the retention.

The retention of volatiles also depends on their *polarity* the more polar, the less retention.[32,65,156] This could be explained by the greater solubility of polar compounds in water. As the water solubility of the volatile increases, the volatile losses increase due to the ability of the water fraction to diffuse through the selective membrane, even at late stages of the drying process. For example, Leahy et al.[155] and Rosenberg et al.[65,67] found that retention of partially water soluble esters (ethyl propionate and ethyl butyrate) in gum Arabic or whey proteins were less than those with lower polarity (ethyl caproate and ethyl caprylate). From their results, they considered that the retention of non polar volatiles is controlled by a combination of molecular diffusivity and droplet stripping because of internal circulation at the early stages of drying.

It should be noted that individual volatiles can be retained at different rates during spray drying encapsulation. Goubet et al.[154] revealed that the retention of aroma

compounds with various functional groups is in the order of acids < aldehydes < esters ≤ ketones ≤ alcohols with acids having the minimum retention. Therefore, it can be seen that retention of volatiles depends on their molecular weight, relative volatility, polarity, and type. These different parameters act on the capacity of the volatile to diffuse through the droplet surface and on its ability to form small pools. The final result is that small, very volatile and water-soluble flavours are lost to a greater extent than the larger, less volatile and water-insoluble flavourings.[19,20,53] Besides factors such as volatility, solubility and diffusivity of the volatile compound through the droplet, another factor that should be taken into account in spray drying microencapsulation is the possible interactions between the volatiles and the wall material.[32] This may involve physical or physicochemical interactions including formation of insoluble complexes, and molecular association of the wall material with the volatile through hydrogen bonds. These interactions can have an effect on the formation of the interfacial film at the interface of O/W, which stabilises the emulsion and may affect the retention indirectly.

Concentration of the Core Material

The common term used in the microencapsulation reports is the core-to-wall ratio, which in fact, is a representative of oil or flavour concentration (load). Using the highest possible core concentration that provides high core retention in the microcapsules is advantageous, because less wall materials will be needed and by increasing the yield and output, it will be better from economic point of view. In general, there is an optimum core concentration that can be encapsulated efficiently. Higher oil loads generally result in poorer retention or lower encapsulation efficiency[46,65,67,89,113,157,158] and higher surface oil content of the powder.[47,69,95,151] For example, a 10% decrease in volatiles total retention and a surprisingly 150% increase in volatiles retained on the surface of powder particles was observed by Bhandari et al.,[69] when the oil load was increased from 20 to 25% of the wall solids. In another work, Hogan et al.[46] found that by increasing soy oil-sodium caseinate ratio from 0.25 to 3.0, microencapsulation efficiency was dramatically decreased from 89.2% to 18.8% respectively during spray drying. This general trend is attributed to greater proportions of core materials, particularly volatiles, close to the drying surface, thereby shortening the diffusion path length to the air/particle interface.

In spite of this trend, in some specific applications, higher volatile loads would also provide higher retention. For example, Sheu and Rosenberg[54] obtained high ethyl caprylate retention in a whey protein/carbohydrate combined wall system for an ester load of 30% (w/w), corresponding to a wall to core ratio of 2.3:1. In most of the published works, a typical core to wall material ratio of 1:4 (20% core at the final encapsulated powder) is usually

adopted and reported as being optimal for various wall materials like gum Arabic and modified starches.[4,7,15,72,97] One of the few exceptions to this is the patent of Brenner et al.[159] who claimed effectively encapsulating up to 75% flavours by the use of a plasticizing wall material (sorbitol) but, there is no commercial products using this patent.[19,20]

Role of the Initial Emulsion

As noted in an earlier section, one of the key steps in spray drying encapsulation of oils and flavours is preparation of the infeed emulsion. This emulsion plays an important role in determining the retention of volatiles and surface oil content of the final encapsulated powder. The significant parameters to consider are total solids concentration, viscosity, stability, droplet size, and emulsification method which are reviewed in this section.

Total Solids Content of the Emulsion

It is shown that the most important factor determining the retention of volatiles and encapsulation efficiency of food oils during spray drying is the dissolved solids content in the feed.[32,116,160–163] High solids content of the prepared emulsion increase retention principally by reducing the required time to form a semi-permeable membrane at the surface of the drying particle. Also higher total solids leads to the increase of emulsion viscosity, preventing the circulation movement inside the droplets and thereby, resulting in a rapid skin formation that will be discussed in the following section.

Although some authors such as Sankarikutty et al.[97] and Rosenberg et al.[65] suggest that the highest possible infeed solids content should be used, later results have shown that there is an optimum infeed solids content for the drying of food flavours and oils.[69,164] Two reasons are mentioned in this regard: first, at some solids content, adding more wall materials exceeds its solubility, and these undissolved wall materials can not provide any effective encapsulating effect and so, leads to poorer flavour retention during the drying process; The second reason for an optimum infeed solid is related to the viscosity of the initial emulsion which is shown to have an optimum figure. The effect of infeed solids content depends on the type of core material. For example, Liu et al.[80] revealed that the retention of d-limonene (more than 95%) was independent of the initial solid concentration, while the retention of ethyl butyrate and ethyl propionate was markedly affected by the solid concentration, similar to the retention of diacetyl in the work of Reineccius and Coulter.[75] Regarding ethyl caproate, Liu et al.[80] found that it was slightly depended on the initial solid concentration. They showed that below 25% solids, particularly, the retention increased steeply with the increase in concentration, possibly due to the rapid formation of crust on the surface of the droplet to
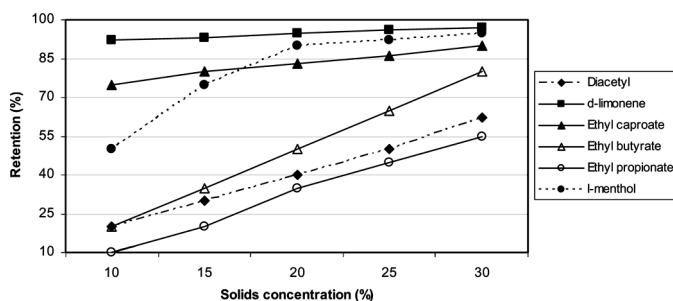
FIG. 4.   Effect of initial solids concentration on the retention of different flavors during spray drying encapsulation. Data from.[72, 75, 80]
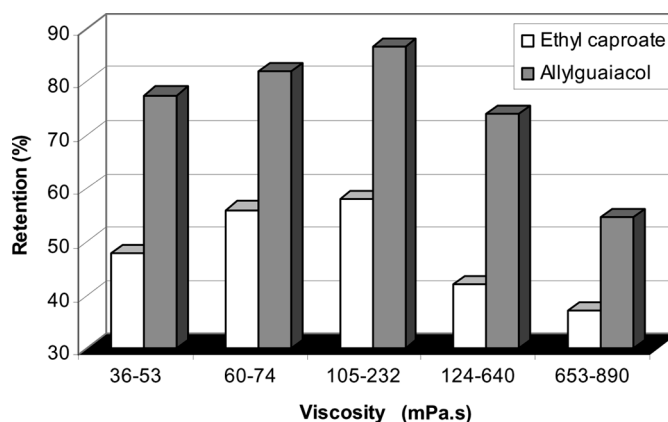

FIG. 5.   Effect of emulsion viscosity through addition of sodium alginate on the retention of ethyl caproate and Allylguaiacol during spray-drying microencapsulation. Data from.[65, 165]

trap volatiles emerging from the ruptured emulsion. Therefore, it can be concluded that infeed solids concentration has a pronounced influence on those volatiles that are most susceptible to loss (low molecular weight), as shown in Figure 4.

Emulsion Viscosity

An increase in the viscosity of the initial emulsion should help volatile retention because of reduction of internal circulations in the droplets and rapid semi-permeable membrane formation. Increasing the solids concentration in the initial emulsion is favourable up to a point that is relevant to optimum viscosity. Some researchers have increased the viscosity of the emulsion without significantly changing its solids content through addition of thickeners (≤1% w/w of wall materials concentration) like carboxyl methyl cellulose, gums, sodium alginate or gelatine. For instance, Rosenberg et al.,[65] and Silva and Re[165] monitored the effect of sodium alginate addition on the retention of ethyl caproate and Allylguaiacol respectively, during spray drying encapsulation. They found an optimum in retention as a function of alginate concentration, which corresponded to an emulsion viscosity ranging from 125 to 250 mPa·s for gum Arabic/ethyl caproate emulsions and about 105 mPa.s for maltodextrin/Allylguaiacol emulsions (Figure 5). They claimed that this viscosity was relatively easy to atomize and reasonably spherical particles were formed. In another study, Liu et al.[99] explained that addition of gelatine at 1% (w/w) markedly enhanced the retention of ethyl butyrate when gum Arabic was used as the emulsifier, because of improved formation of crust on the surface of the droplets. They found no appreciable change in retention when soy soluble polysaccharides (SSPS) were used as the emulsifier. Reineccius and Coulter[75] reported similar results (no significant improvement in flavour retention) when they increased the infeed viscosity through the addition of xanthan gum during drying.

To summarize, it is obvious that emulsion viscosity plays an important role in determining volatiles retention,

due to its large influence on the control of volatile losses until the surface of the drying droplet becomes semi-permeable. In other words, increasing the emulsion viscosity up to an optimum point will suppress the internal circulations and oscillations of droplets, and will put the selective diffusion into action earlier, so improves retention. However, increasing the viscosity beyond that optimum limit causes a decrease of the retention, due to a larger exposure during atomization, the slow formation of discrete droplets during atomization, and difficulties in droplet formation. It is shown that a more viscous feed will produce larger droplet sizes, and due to difficulties on droplet formation at higher viscosities, irregular particles (oval, cylindrical, and stringy) will be produced.[32,69,165,166]

Emulsion Stability

The encapsulation efficiency of oils and flavours is expected to be influenced by the stability of initial emulsion: better the stability, higher the efficiency.[67,88,117,118] For example, Hogan et al.[46,89,145] showed that micro-encapsulation efficiency of soy oil with milk proteins and carbohydrate blends was negatively correlated with emulsion size of the reconstituted spray dried powders, which is a representative of emulsion stability during the process. Also, in a series of works on single droplet by Liu et al.,[80,99,167] they studied the effect of emulsion stability on the retention of emulsified hydrophobic flavours during drying. As a measure of stability, they examined the decreasing rate of emulsion absorbance,[80] or the increase of emulsion droplet size against time.[99] In their earlier work,[167] the time course of emulsion droplet size was used and they found that the natural log of mean droplet diameter was linearly proportional to the natural log of time, for both d-limonene and ethyl butyrate. For each flavour, the final retention correlated negatively with the emulsion

stability, demonstrating that unstable emulsion was broken inside the droplet, resulting in appreciable loss of flavour during drying. Their results also showed that ethyl butyrate emulsion size was growing extremely rapid (about 20 to 40 times faster) compared with d-limonene, implying that d-limonene emulsion was much more stable than that of ethyl butyrate, resulting in a higher retention during drying. Another important result was that the viscosity of an unstable emulsion such as ethyl butyrate-gum Arabic emulsion was higher and changed greater than a stable emulsion. Since the emulsion droplets of ethyl butyrate would break down either inside or on the surface of sprayed droplets, its loss during drying was higher than that of d-limonene. The same results were also reported in another study by Soottitantawat et al.[60] who showed that retention of d-limonene (80 to 95%) was higher than the esters, ethyl butyrate (40 to 60%) and ethyl propionate (40 to 50%).

Finally, Liu et al.[99] found that by adjusting the density of the ester flavours with a weighting agent, sucrose acetate isobutyrate, the emulsion stability and flavour retention can be improved. However, the retention of these density adjusted ester flavours during spray drying was still lower than the retention of the stable emulsion of d-limonene, indicating that some other factors such as the molecular weight, volatility and solubility of the flavours possibly influence the retention, as discussed before in section 4.2.

Emulsion Size

Quite apart from emulsion viscosity and stability, some workers have shown that emulsion size has a considerable effect on the encapsulation efficiency of oils and flavours during spray drying microencapsulation.[54,59,60,70,72,99,117,167,168] In each of the reported studies, the encapsulation efficiency of a particular core material improved with decreased emulsion droplet size. In approximately all of the cited studies, emulsion size has been decreased to about one micron, and there is no indication of reducing the emulsion size further to submicron range to know whether submicron emulsions can improve the encapsulation efficiency or not? To obtain an emulsion with fine droplets ($< 1.0\,\mu m$), a favourable surface-active biopolymer with appropriate emulsification method should be employed.

One advantage of producing a finer emulsion is higher stability, which is critical during spray drying. The emulsion size may also affect the characteristics of the final encapsulated powder including the surface oil and total oil content of the microcapsules. For example, Risch and Reineccius[168] by reducing the emulsion size to the minimum ($0.90\,\mu m$) through Microfluidization, found that a smaller emulsion size yielded a higher retention and lower unencapsulated oil (surface oil) on the dried powders of gum Arabic/or modified starch and orange oil emulsions,

but surprisingly powders with larger emulsion sizes had a longer shelf life. However, Minemoto et al.[117] revealed that encapsulated linoleic acid with smaller emulsion size, oxidizes more slowly than powders with bigger emulsion sizes, possibly because of lower amounts of unencapsulated oil on the surface of spray dried particles. Similar results were obtained by Liu et al.,[167] and recently Soottitantawat et al.,[59,60,72] who showed that for different wall materials (e.g., gum Arabic, Hi-Cap, and maltodextrin), the increasing emulsion oil diameter resulted in a decreased retention of d-limonene, that was more pronounced in the finer emulsions (less than $2\,\mu m$), as presented in Figure 6. This implies that a fine emulsion is stable during both the atomization and spray drying processes, and the emulsion droplet size is a significant factor for the retention of flavours. Their results also showed that the powder size was not affected by changing the emulsion size.

In contrast to these reports, Rosenberg and Sheu[67] found that the retention of ethyl caprylate in whey protein isolate/lactose was higher than that in whey protein isolate although the latter resulted in a smaller emulsion droplet size. They hypothesized that the effect of lactose on drying and crust formation (and hence on ethyl caprylate retention) was more significant than the effect of emulsion size distribution. Also, Hogan et al.[46] revealed that microencapsulation efficiency of soy oil encapsulated powders with sodium caseinate was not affected by the homogenization pressure, which corresponded to emulsions with different sizes. Regarding more water soluble flavours (ethyl butyrate and ethyl propionate), Soottitantawat et al.[60] observed different behaviour than d-limonene (more lipophilic): there appeared to be an optimum emulsion size for the retention of these volatiles, i.e., esters (Figure 6). They explained that the increased loss of the esters at small
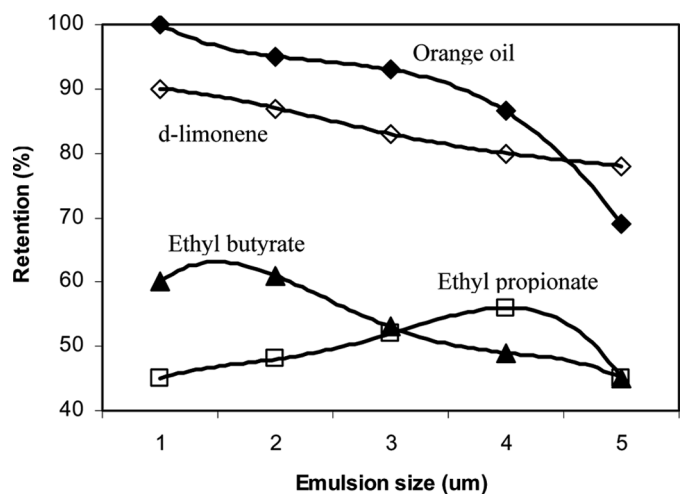

FIG. 6.   Influence of emulsion droplet size on the retention of flavors during spray-drying encapsulation. Data from.[60, 168]

droplet sizes could be due to the larger surface areas of the fine emulsions, which present a greater opportunity for diffusion into the matrix and loss from its surface during drying. Again at larger emulsion sizes, the droplets would be subject to shear losses. Soottitantawat et al.[60] also provided data showing that atomization resulted in a decrease in droplet size of the coarser emulsion but had no effect on the finer emulsion. They claimed that the shearing effect during atomization disrupts larger emulsion droplets allowing them to evaporate during drying, contributing to the greater loss of flavours from the larger emulsion droplets during spray drying.

Another important result of the work of Soottitantawat et al.[60] was that the amount of surface oil increased with the increasing emulsion droplet size (Figure 7), in agreement with the results of Risch and Reineccius,[168] and Danviriyakul et al.[88] Higher remaining oil on the surface of encapsulated particles might be explained by the breakdown of the large emulsion droplets during atomization, and inefficient encapsulation of big oil droplets. The oil on the surface of the dried microcapsules is very important for stable storage, because it has no protection against oxidation, and can be easily oxidized to form off-flavour compounds. Therefore, finer emulsions may contribute to keep the core material inside the particles within acceptable levels for a longer period of time, although this does not necessarily correspond to a longer shelf-life, or a higher resistance to oxidation in the product, since the greater surface area of the oil droplets embedded in the capsule wall provides greater possibility for oxidation once oxygen has penetrated into the particle.[19,20]

Finally, in a study by Re and Liu,[70] they found that retention of volatiles (Eugenol) was directly related to the difference between emulsion droplet size and particle size of the spray dried microcapsules (Figure 8). Therefore, it
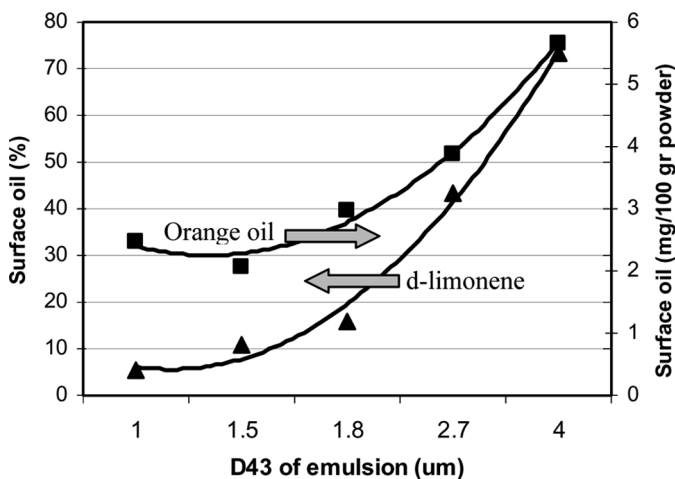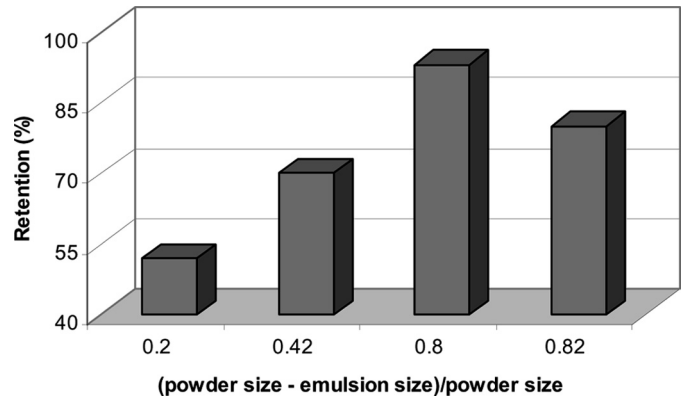
FIG. 8. Influence of the difference between the emulsion droplet size and the particle size of dried microcapsules on the volatile retention. Data from.[70]

could be possible to improve the retention by increasing the difference between emulsion size and powder particle size. This might be explained by more efficient covering of fine oil droplets inside the wall material and minimum effect of atomization and spray drying on emulsion droplets. However, more works need to be done in this area to find the exact mechanism of the influence of emulsion size and powder particle size on encapsulation efficiency of oils and flavours during spray drying.

Emulsification Method

While the emulsion size is only one factor which can influence the characteristics of the spray dried microcapsules, it may be possible to use this parameter in combination with other data and information (e.g., emulsification method) to manufacture a product with better emulsion stability, an extended shelf-life, and higher flavour load. In an investigation by Mongenot et al.,[96] their results have clearly shown that the use of ultrasound increases emulsion quality when the wall material has low emulsifying properties and a weak viscosity, such as Maltodextrin, resulting in a higher aroma retention than the use of Ultra-Turrax and permits limited diffusion of the most volatile and polar compound during drying. They observed an increase of aroma retention when using esterified modified starch as the wall material with ultrasonic emulsification, whereas no significant difference in emulsion size existed between Ultra-Turrax and ultrasound. Mongenot et al.[96] also reported a weak retention of butyric acid when Ultra-Turrax was the emulsification technique, compared with a high retention when ultrasonication was used. On the other hand, they found no significant difference in the retention of lemon aroma with modified starch, between the two emulsification methods. They concluded that for all samples, in general, the use of ultrasound emulsification resulted in the strong retention of butyric acid

FIG. 7. Effect of initial emulsion droplet size on the amount of surface oil on the encapsulated powder after spray drying. Data from.[60, 185]

(the most polar and volatile compound), while a weak retention was observed for lauric acid (the less polar and volatile compound). Although emulsion size can be one factor, but other properties including emulsion size distribution and powder size could make the difference between two emulsification methods, and should be considered.

Conditions of the Spray-Drying Process

If the infeed emulsion is stable enough with optimum conditions such as viscosity and droplet size, encapsulation efficiency could be maximized by the right choice of spray drying parameters including inlet and outlet drying air temperatures, infeed temperature, atomization type and conditions, drying air flow rate and humidity, and powder particle size.

Powder Particle Size

Particle size of the encapsulated powder is primarily determined by the physical properties of the emulsion to be dried (such as viscosity and solids concentration), and the operational parameters chosen for atomization, such as the rotational speed and wheel diameter in the case of centrifugal wheel atomization, and the orifice size and pressure in the case of nozzle atomization.[19,20,53,62,63,64,115] For instance, a high pressure and small orifice will result in smaller particles. Finney et al.[64] found that nozzle atomization produces substantially bigger particles than centrifugal wheel atomization, and the type of atomization was evidently more important in determining particle size distribution than dryer temperatures. Particle size can also be influenced by the operating temperatures: high inlet air temperature and low difference between inlet and exit air temperatures will produce slightly larger particles than drying under conditions that result in slow drying. This is due to the fact that very fast drying sets up the particle structure early and does not allow the particles to shrink during drying. Infeed solids have a similar effect in that the particles dry quickly if they are high in solids and can not shrink as much.[19,20] In practice, depending on the spray dryer design, it is possible to control particle size to some extent, based on the mentioned parameters.

The influence of powder particle size on encapsulation efficiency of food favours and oils has not been clear. Several workers have reported that larger particle sizes result in improved flavour retention and lower surface oil contents during spray drying.[66,115,169] On the other hand, Reineccius and Coulter[75] and Finney et al.[64] could find no effect of particle size on retention, as they attributed this result to the high concentration of infeed solids, i.e., particle size is not important if high infeed solids are used. These controversial data can be mainly related to variations in the spray drying design and methods to control particle size, or the properties of the initial emulsion. Recently, Soottitantawat et al.[72] showed that powder particle size alone does not have a significant effect on flavour retention, as other parameters such as emulsion size can have a more considerable influence (Table 2). They concluded that larger powder size leads to higher stability and lower release of encapsulated flavour, if the initial emulsion has a small size. Furthermore, Zakarian and King[170] have shown that if both volatile loss and rate of drying are diffusion controlled, volatile retention should be independent of particle size.

The work of Silva and Re[165] suggested the existence of an optimal particle size to achieve maximal volatile retention, similar to the results of Chang et al.,[171] who found that the total oil retention was highest for powder with intermediate particles, while it was lowest for powder with largest particles (Table 2). They produced encapsulated orange oil powders with three different particle sizes through varying the voltage supply of a centrifugal wheel atomizer during spray drying. The other result was that powders with medium and large particle sizes had about 2.5 and 9 times more surface oil than powder with small particle size, respectively (Table 2). This was not expected since powders with larger particles have less surface area and therefore, should have less surface oil, similar to the findings of Finney et al.[64] These workers explained that while large particles have a reduced surface area to volume ratio, which would result in better core retention, there would be also a longer time for film formation around the large droplets during the process. The longer the time necessary for film formation, the greater the loss of volatile substances. These two competing factors will thus determine the overall effect of particle size upon volatile retention. Another reason for poorer volatile retention and higher surface oil content observed for larger particles could be related to the surface morphology. It has been well documented that when there is a slow process of film formation around the droplets, the resulted particle will have surface imperfections. In fact, the damage of the particles surface integrity (fissures, shrinkage) observed mainly for larger particles, which result in an increase of their surface area, may contribute to the increase of the unencapsulated or surface oil.

Recently, Jafari et al.[172] investigated the role of powder particle size by classifying an spray dried encapsulated powder into three different ranges with vibrated screens. They reported particles with medium size have the highest encapsulation efficiency. Although the role of particle size is not clear, it is often desirable to produce large particles to facilitate rehydration. Small particles tend to disperse very poorly, especially in cold water, and instead form lumps on the liquid surface. Large particles can be obtained through appropriate choice of spray dryer operating conditions, or the use of agglomeration techniques.[74,173,174] These recent works have shown that agglomeration of spray dried encapsulated powders through fluidized bed processing

TABLE 2
The influence of particle size of the encapsulated powders on encapsulation efficiency during spray drying

Encapsulation ingredients	Particle size control via	Powder particle size (µm)	Retention (%)	Surface oil content (%)	Reference
Modified starch (30% solids) + orange oil (20% of solids)	Change in centrifugal atomizer voltage	42.5	15.2	0.67	[171]
		53.2	15.9	1.68	
		66.6	12.8	7.10	
Maltodextrin and soy lecithin (40% solids) + Allylguaiacol (25% of solids)	Change in emulsion viscosity	2.4–26.2	77.5		[165]
		2.4–29.4	82.3		
		2.9–31.1	86.6		
		3.1–35.9	74.2		
		2.9–46.9	69.8		
		3.4–64.0	54.7		
Modified starch (40% solids) + orange oil (25% of solids)	Change in atomizer type and temperature	35.1	97.5	16.5 (mg/100 gr powder)	[64]
		40.8	99.9	13.2	
		65.1	99.9	8.5	
		76.7	97.5	5.3	
Modified starch (20% solids) + d-limonene (25% of solids)	Change in rotational speed of atomizer	25–30	80–89	0.46–1.28 (based on emulsion size)	[72]
		40–50	74–92	0.39–0.63	
		60–70	79–94	0.43–1.25	

can improve the flowability and wettability of powders through increase in particle size to about 200 µm. Some of the encapsulation parameters could also be changed, such as reduction in surface oil content of the powders due to stripping effect of fluidized bed agglomeration.[173]

Atomization Type

As mentioned before, significant losses of volatiles occur during early stages of drying, particularly in the atomization step. During this time, the emulsion is sprayed into very turbulent air, forming a thin sheet (high surface area) with substantial mixing, all enhancing volatile losses. Thus, it is necessary to optimize the atomization process for maximum volatile retention. In the case of pressure nozzles, it is shown that a higher pressure enhances volatile retention. For example, King[81] found that atomization at 1.83, 3.55 and 7.00 MPa resulted in 31, 45 and 53% retention of propyl acetate during drying, partially due to reducing the length of the emulsion sheet emitted from the nozzle atomizer before break-up into spherical droplets, thereby reducing the length of time that the liquid is in a sheet (high loss rate period). He explained that also high pressures provide a greater momentum to the atomized droplets, thereby drawing more hot air into the spray stream, so more rapid drying and quicker formation of the selective film around the drying droplet. Another

parameter according to King[81] is the spray angle of the nozzle that can affect volatile retention: a wide spray pattern (without wetting the dryer walls) is recommended, since this will increase the atomized droplet contact with drying air, thereby increasing the drying rate. He concluded that the same situation exists for centrifugal atomization: higher wheel speed would enhance volatile retention for similar reasons. Recently, the results of a work by Finney et al.[64] showed that neither type of atomization nor processing temperatures had a significant influence on the retention of orange oil, with overall excellent flavour retention (Table 2). Also, powders produced by centrifugal atomization had much higher surface oil contents than their nozzle counterparts. So, the type of the atomization process and the associated dryer geometry can influence the encapsulation efficiency of food oils and flavours, an effect indirectly related to the powder particle size.

Infeed Temperature

This parameter has also been studied by many workers.[160,169,175–178] For example, Sivetz and Foote[175] found that cooling the feed (30% coffee solids extract) before drying markedly improved the coffee flavour of the final spray dried powder, possibly due to an increase in the feed viscosity, which, in turn, would affect internal circulations of the droplets and size of the atomized droplets, along

with the vapour pressure and diffusibility of the flavour compounds.[19,20] Also, Thijssen[176–178] stated that infeed temperature should be increased to the point that higher infeed solids (i.e. greater solubility) may be used, resulting in better retention mainly because of higher solids. One problem with higher infeed temperatures could be microbial growth in these materials before drying.[19]

Air Flows and Humidity in the Spray Dryer

The better the mixing of air and atomised emulsion, the better is the retention of volatiles, due to a more rapid heat and mass transfer associated with the drying process, i.e. more rapid drying[161]. This parameter is primarily determined by spray dryer design, and so can be changed only to a limited degree[62, 63]. For example, establishing a wide spray pattern with nozzle atomization or higher pressures will improve air/product mixing thereby, enhancing the drying rate, as reviewed earlier. Lower dryer air humidity can also promote rapid drying and better flavour retention, but dehumidifying the inlet air typically is expensive and, is rarely performed during spray drying encapsulation.[19,20,53]

Inlet Air Temperature

The influence of spray dryer inlet and outlet air temperatures on encapsulation efficiency of food oils and flavours has also been the subject of numerous studies.[33,56,61,65,69,80,179,180] It is shown that a high enough inlet air temperature (160–220°C) leads to a rapid formation of the semi-permeable membrane on the droplet surface, giving optimum flavour retention beyond which, could cause heat damage to the dry product, or "ballooning" and excessive bubble growth and surface imperfections which increase losses during spray drying.[108,160,169,176] Ballooning occurs when steam is formed in the interior of the drying droplet due to quite high inlet air temperatures, causing the droplet to puff (or balloon), thereby producing a thin-walled hollow particle. This particle will not retain core materials as well as its non-ballooned counterpart. The ballooning temperature is shown to be mainly a function of the used wall material and spray dryer design, and spray dried encapsulated volatiles have been successfully produced using inlet air temperatures up to 280–350°C.

For instance, Bhandari et al.[69] observed up to 84% volatile retention with a tendency to increase at higher inlet air temperatures up to 400°C, using a leaflash spray dryer system without any serious "ballooning" at a reasonable exit air temperature. Shiga et al.[61] also reported higher retention of shiitake flavour encapsulated with cyclodextrins and maltodextrin at higher drying temperatures On the other hand, Reineccius and Coulter,[75] Anker and Reineccius,[179] and Aburto et al.[180] showed that the retention (total oil) of diacetyl and orange oil, was independent of

the air temperature, similar to the study of Finney et al.,[64] who presented data confirming that even the surface oil increased by increasing the air temperature. While Bhandari et al.[69] found a decrease in surface oil content of powder particles by increasing the inlet air temperature, possibly because of rapid drying rate that would make the membrane around particles firmer, and no further leaching of the volatile could occur towards the surface. Anker and Reineccius[179] found similar results regarding surface oil content and concluded that the powder dried at the highest operating temperature (280°C inlet air) had the maximum shelf life, since surface oil decreased with increasing inlet and outlet air temperature differential.

Considering different volatiles, Liu et al.[80] showed that retention of d-limonene was independent of air temperature, while the retention of ethyl caproate slightly increased as the air temperature increased from 40 to 100°C, similar to the results of Rosenberg et al.,[65] who revealed that the influence of inlet air temperature on retention of ethyl caproate was stronger at higher solids concentration. Liu et al.[80] claimed that for d-limonene or ethyl caproate (low soluble), the emulsion is stable so the retention is high and independent of hot air temperature. On the other hand, the emulsions of ethyl butyrate or ethyl propionate are so unstable that could break inside the droplet during drying, and the emerging flavour may diffuse through the droplet surface. However, when the air temperature is high, the crust formation is quick and the flavour can not evaporate easily from the surface. Liu et al.[80] explained that at the air temperature of about 115°C, the droplet experiences a morphological change of cyclic inflation and bursting for ethyl butyrate and ethyl propionate and their retention decreases abruptly, because of the probable breakdown of these flavour emulsions on inflation or bursting of droplet, and exceeding the droplet temperature above their boiling point.

Outlet Air Temperature

The influence of outlet air temperature on the encapsulation efficiency of food flavours and oils is also controversial and unclear. For example, Reineccius and Coulter[75] found that retention of small soluble volatiles such as diacetyl improves with increasing outlet air temperatures, probably due to a lower relative humidity at higher outlet air temperatures (at a fixed inlet air temperature), which results in more rapid drying as discussed before and therefore, better flavours retention. In contrast, Bhandari et al.[69] found that increasing outlet air temperature results in poorer volatiles retention and higher surface oil contents of particles. They hypothesized this by "ballooning" effect, where particles may develop fissures, even split and release the trapped volatiles. This was validated by their data showing decrease in particle density of the powder. A similar trend was reported by Anker and

Reineccius[179] who found in a constant inlet air temperature, the surface oil increases at higher outlet air temperatures. But, they didn't observe any significant change in orange peel oil retention by increasing the exit air temperature. Recently, Danviriyakul et al.[88] also revealed that surface oil content of milk fat encapsulated powders was not affected by outlet temperature.

CONCLUSION

Currently, the main emphasis of the microencapsulation of food oils and flavours has concentrated on improving the encapsulation efficiency during spray drying and extending the shelf-life of the products. This is intended to produce high quality encapsulated powders. The properties of the wall and core materials as well as the emulsion characteristics and drying parameters are the factors that can affect the efficiency of encapsulation. The infeed emulsion size plays an important role on the retention and the stability of the encapsulated oils and flavours. Previous results have indicated that there are advantages to create a smaller emulsion size for the insoluble flavours (such as d-limonene) and an optimal value for moderately soluble flavours such as ethyl butyrate and ethyl propionate. The key advantage of smaller emulsion sizes is a better retention of volatiles in the spray dried powder. This results in a direct economic benefit to the manufacturer and user of the product. Less flavour is lost during drying and less powder, therefore, is needed in the finished product to achieve the same flavour level. A second advantage is that smaller emulsion sizes also yield dried powders which have less extractable surface oil. As mentioned before, the oil on the surface has no protection from oxidation. A larger amount of extractable surface oil that can readily oxidized, could give a dry product, such as a fish oil encapsulated powder, an off-flavour. A third advantage of producing a finer emulsion is that the emulsion is more stable. This is particularly important in beverage applications where viscosity can not be increased to help stabilize the flavour emulsion. These are the three distinct advantages of creating finer emulsions for spray drying microencapsulation.

The role of particle size of the atomized droplets in determining flavour retention has been controversial. Several workers have reported that larger particle sizes result in improved flavour retention during spray drying. In contrast, some other workers found no effect of particle size on retention. Further, several studies suggested the existence of an optimal particle size to achieve maximal volatile retention. Importantly, it has been shown that flavour retention improved as the difference between the mean emulsion size and the mean particle size increased. Finally, there have been results indicating that emulsification itself can improve flavour retention during spray drying microencapsulation. However, there is no clear cut on how the flavour and oil droplets are retained in various size of powder particles and whether the distribution of the emulsion is the same in every powder particles produced by spray drying. How can the type of emulsification process influence the encapsulation efficiency of food flavours and oils, even with the same emulsion size? What is the effect of emulsion size in nano range? How can nano-emulsions along with nano-particle encapsulation improve the retention and stability of flavours and oils during spray drying? Is there any difference for volatiles and non-volatiles by considering emulsion size and powder particle size? There are insufficient studies being reported on the relationship between the emulsion size the powder particle size and their effect on the efficiency of encapsulation. Commercially, several encapsulated food powders containing active compounds are produced and marketed. However, the presence of high amounts of surface oil is a big draw-back for the process. Even small amounts of highly oxidisable material will quickly deteriorate whole product.

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