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Encapsulation of colorants by natural polymers for food applications

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Product appearance is an important factor for consumers when determining the quality of a product, and colour is one of the most important factors which contribute to product appearance. Currently, the safety and consumer acceptance of some colorants used in food products, such as titanium dioxide and some synthetic colorants, are under discussion. Therefore, new ways to use natural colorants as alternatives to these suspect colorants for future applications are being investigated. A promising method for increasing the applicability of the often sensitive natural colorants is the encapsulation of these colorants in colloidal particles by natural polymers such as carbohydrates, lipids and proteins. In recent years, micro- and nano-encapsulation have increasingly been used for various purposes concerning several food properties such as colour, flavour and micronutrient content. This technique results in improved stability for the often sensitive natural colorants and presents the possibility of entrapping water-insoluble colorants for improved use in an aqueous system. This paper reviews the main methods that are used for encapsulation by natural polymers, discusses the different types thereof that are used for encapsulation of colorants, and provides a short overview of natural colorants successfully encapsulated in these natural polymers.

Introduction

It is widely known that product appearance is one of the primary factors that determines the perceived quality of a product by consumers. Colour is one of the most important appearance factors used by consumers to gain a first impression and judge whether the product meets their standards or not [1–3]. Nowadays, consumers are increasingly aware of what they are eating, which is represented by the famous adage: 'You are what you eat'. Some consumers express a desire for their food products to be natural and that they should only contain ingredients which are, in their opinion, 'natural'. This has resulted in an increase in the research directed towards natural food ingredients and colorants [4,5]. A similar effect is observed in the research undertaken towards the safety and possible negative effects of inorganic and synthetic food colorants. The most well-known and widely used colorant is titanium dioxide, which accounts for 70% of the total production volume of pigments worldwide [6]. Titanium dioxide is used as a pigment in many industries, primarily in paints and coatings. Additionally, it is also used as a pigment in plastics, paper, inks, textiles, photovoltaic cells, biomedical devices, cosmetics, and even as a food additive [7,8]. In the food industry it is used as a white colorant and opacifier; it is authorised as a food colorant in the European Union (EU) as E171. Although titanium dioxide is considered food-safe, concerns have been raised on the safety of titanium dioxide nanoparticles that were also found in food products [9,10]. The International Union of Pure and Applied Chemistry (IUPAC) definition of a nanoparticle is a particle of any shape in the 1–100 nm range, whereas a microparticle is defined as a particle with dimensions of 0.1–100 μm [11]. Recently, the oral intake of titanium dioxide via food and toothpaste was determined based on an average lifespan of 80 years. It was found that the lifelong daily intake of titanium dioxide for people in The Netherlands was 0.19 mg/kg of bodyweight, of which 0.62 μg/kg of bodyweight was titanium dioxide particles [12]. Using this information, toxicological experiments were performed indicating it is possible that there are negative long-term health effects associated with the oral intake of titanium dioxide nanoparticles. However, additional research on this topic is necessary to provide a good overview of the potential long-term negative effects associated with titanium dioxide nanoparticles [13,14].

Not only inorganic colour additives such as titanium dioxide are under discussion, but also some synthetic colour additives. Here, questions are raised mainly concerning reported negative behavioural changes in children, as evidenced by behavioural disorders such as attention deficit hyperactivity disorder (ADHD). The colorants in question are Sunset Yellow FCF (E110), carmoisine (E122), tartrazine (E102), Ponceau 4R (E124), Quinoline Yellow WS (E104) and Allura Red AC (E129). Parents have reported behavioural changes in the form of hyperactivity after their children have consumed these synthetic colorants [15]. In a later study [16] by the same group, the same behavioural changes were observed using standardised clinical tests. The researchers concluded the following: 'Artificial colors or a sodium benzoate preservative (or both) in the diet result in increased hyperactivity in 3-year old and 8/9-year-old children in the general population'. However, many aspects are still unclear because of inconsistencies in different research methods into this topic and the possibility that genetic or environmental factors could also influence these

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outcomes [17]. For example, the same experiment was repeated in Hong Kong [18], where the researchers concluded that there were no significant associations between either artificial food colorants or a preservative on Chinese children's behaviour at the age of 8–9 years. Overall, the results from multiple studies on negative behavioural changes and toxicological effects are inconclusive, and research into this topic is ongoing [19].

These inconclusive data on the safety and negative effects of inorganic and synthetic colorants are, at the present time, not particularly helpful for consumers. Another reason for consumers not to trust non-natural colorants is the fact that several previously approved food additives, initially believed to be safe to use, are no longer permitted because of proven adverse effects such as toxicity in the medium and long term, as well as a high frequency of health disturbance incidents. [20]. This has motivated the search for and development of naturally derived and safer alternatives for these now questioned synthetic colorants. Recently, Coultate and Blackburn [4] wrote an extensive review on this topic, discussing the past, present and future of food colorants. These natural colorants are plant-, animal-, microbial- and mineral-based. Some commonly used and more prevalent natural colorants and additives include anthocyanins, betalains, carminic acid, carotenoids, chlorophylls, curcuminoids, minerals, phycocyanins, and some vitamins [4]. A disadvantage of using natural colorants is that applications can be limited by their properties. Natural colorants often have a weaker tinctorial strength than their synthetic counterparts, and it is often difficult to match desired hues, especially green and blue. Most natural colorants are sensitive to heat, light and oxygen, which may result in colour loss or shifts. Others may be sensitive to food matrix conditions, such as pH, proteins, metal ions, or some organic compounds [5].

In this review, we discuss encapsulation of these natural colorants by natural polymers, also called biopolymers, for the formation of colloidal particles. Encapsulation offers improved stability for the often sensitive natural colorants by creating a barrier. An additional benefit of this is that possible off-flavours of the colorants can be masked by the biopolymer [21,22]. Also, stability can be further improved by adding antioxidants and other stabilisers to the particles. Using encapsulation for the delivery of colorants presents the possibility of entrapping water-insoluble colorants and using them in an aqueous system [23], which broadens the number of natural colorants that can be easily used in food systems. The encapsulation method has various applications for the purpose of encapsulating colorants, not only in the food industry, but also in the pharmaceutical, cosmetic, textile, and other industries.

Commonly used methods for encapsulation using natural polymers

Encapsulation is defined as the technology of packaging solids, liquids or gaseous materials in matrices (encapsulants) that can sustain and possibly release their contents under specific conditions. Encapsulation can have multiple purposes, the first of which is to protect sensitive components from light, moisture, or heat. Second, encapsulation can be used for improving the stability of biological pigments as a dispersion in aqueous systems to increase the usability of natural colorants in many industrial applications. Another advantage of using encapsulated

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products is that their protective shell stays intact and therefore the colorants do not easily migrate through the product as conventional colorants do. The brightness of the colorants can also be improved, thus increasing their marketability for commercial use [24]. Encapsulation can be achieved using various processes. Examples include emulsification, coacervation, molecular/inclusion complexation, anti-solvent precipitation, emulsification-solvent evaporation technique, supercritical fluid technique, spray-drying, electrospraying, and freeze-drying [25,26]. Commonly encountered encapsulation techniques for the purpose of encapsulating colorants are spray-drying, electrospraying and anti-solvent precipitation. A distinction can be made between encapsulation resulting in dry particles or particles in a liquid: spray-drying and electrospinning yield dry particles, while anti-solvent precipitation yields particles that are dispersed in a liquid. However, anti-solvent precipitation can also yield dry particles by adding a drying step to the procedure, often spray-drying or freeze-drying. Next, the principles of these techniques are explained, including the advantages and challenges of using these techniques in the encapsulation of natural colorants by biopolymers; the possibilities of scaling up procedures for future industrial applications are also explored.

Spray-drying

Spray-drying is the most commonly used encapsulation technique for food products. The spray-drying technique is based on the transformation of a fluid into dry powder by spraying the liquid in a drying gas stream. Figure 1 shows a typical spray-drying set-up. First, the liquid is sprayed into a drying chamber through a nozzle or atomiser. The small droplets that are generated are subjected to fast solvent evaporation, which leads to the formation of dry particles that are separated from the drying gas by a cyclone that deposes them in a collector [27]. Inclusion of an encapsulant in the solution results in the encapsulation of the active component, the encapsulate. The application of the spray-drying process in microencapsulation involves three basic steps [28]: (i) preparation of the dispersion or emulsion to be processed; (ii) homogenisation of the dispersion; and (iii) spraying of the mass into the drying chamber. By selection of the encapsulant, different properties can be achieved. For example, the solubility of the encapsulant directly determines the behaviour when introduced into an aqueous environment, either releasing the core material or retaining its encapsulated state.

Because the spray-drying technique is relatively old (on an industrial scale it dates back to the 1920s), there are many variations in its set-up [29]. The final product quality depends on the operating conditions, such as inlet and outlet air temperatures, feed flow rate, and spraying speed and pressure [29,30]. The particle sizes obtained are often in the microparticle range (0.1–100 μm) [27], which can be a disadvantage when aiming for nanoparticles (1–100 nm). Also, the relatively high operating temperatures during the drying process can affect the stability of thermally sensitive ingredients [31].

The spray-drying technique is already widely used in industry because it is simple and relatively inexpensive, it rapidly and continuously produces dry powder (compared to freeze-drying), and the particle size distribution can be controlled. An additional advantage is that this technique is less dependent on the solubility of the additive and the polymer than other methods [24,32]. In addition, this technique can be used as a final step to remove the solvent to produce dry powder from particles in a liquid medium that were synthesised via a different method.

Electrospraying

Electrospraying is an electrohydrodynamic process, one where a biopolymer solution can be sprayed by the application of a high potential electric field to obtain particles. This technique is closely related to electrospinning, from which fibres and not particles are obtained. A typical electrospraying set-up consists of four main

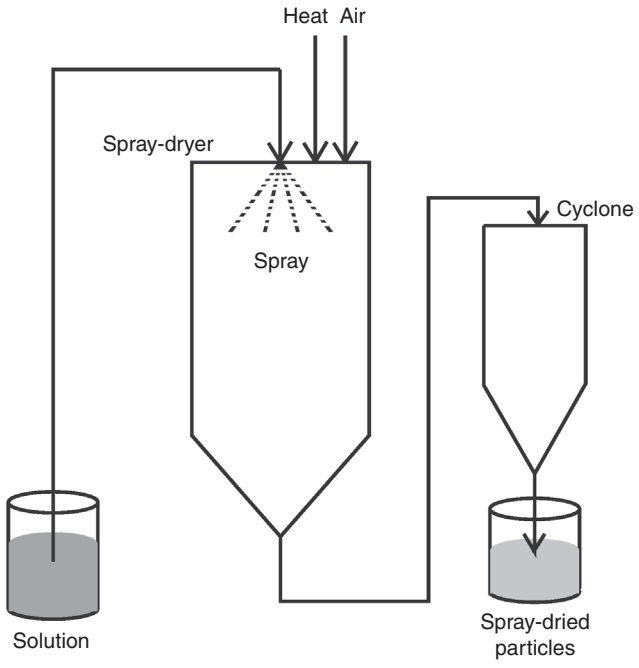

Figure 1 Schematic overview of a spray-drying process

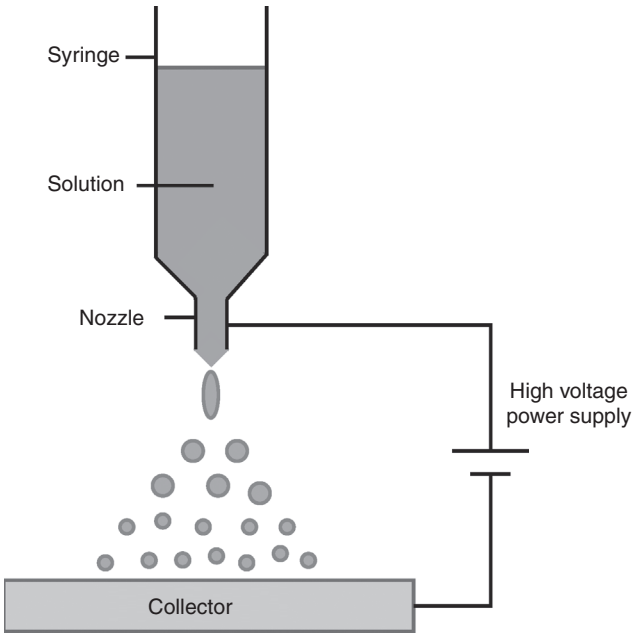

Figure 2 Schematic overview of an electrospraying process

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components: a high voltage power supply, a blunt needle or capillary, a syringe pump, and a grounded collector plate (see Figure 2 for a schematic overview). The main variables that affect the electrospraying process include the properties of the polymer in solution (concentration, molecular weight, viscosity, surface tension and conductivity of the solvent), and the processing conditions (flow rate, applied voltage, and distance between the tip and the collector plate). At high polymer concentration, fibres are formed (electrospinning), and at low polymer concentration, particles are formed (electrospraying) [33]. These variables can be fine-tuned to synthesise particles in narrow particle size distributions at micro (0.1–100 μm) or nano (1–100 nm) scale.

Encapsulation using electrospraying can be achieved using four different main procedures [34]. The first is the collision of two oppositely charged droplets. In this procedure, there are two capillary nozzles with opposite potentials, and the sprayed droplets collide and form a capsule by enveloping the droplet of higher surface tension within the droplet of smaller surface tension. The second procedure is electrospraying and evaporation of colloidal suspension, in which a suspension is electrosprayed and a shell is formed by solvent evaporation. The third is electrospraying and gelatinisation of a colloidal suspension. In this procedure, the core material is electrosprayed into a bath with a gelatinising agent, which forms a hard shell around the core material. The fourth procedure is electro-coextrusion. Here, two different liquids are simultaneously sprayed from two coaxial capillaries at the same potential. The core liquid flows from the central capillary and the encapsulating liquid from the annular nozzle surrounding the central capillary. As with spray-drying, through selection of the encapsulant, different properties can be achieved. For example, the solubility of the encapsulant directly determines the behaviour when introduced into an aqueous environment, either releasing the core material or retaining its encapsulated state.

An advantage of electrospraying is that particle aggregation can be prevented because of the charge the droplets develop in the spraying process [35]. Moreover, the electrospraying technique is an easy one-step process for the production of dried nanoparticles. This is opposed to spray-drying where mostly microparticles are obtained, since the particle collection method of a conventional spray-drying set-up needs to be replaced to obtain nanoparticles [36]. Electrospraying can also be advantageous compared with anti-solvent precipitation (discussed further in the Anti-solvent precipitation section), where the goal is to obtain dry particles. Because the particles are dispersed in a liquid medium in anti-solvent precipitation, an additional drying step is necessary to obtain dry particles [37].

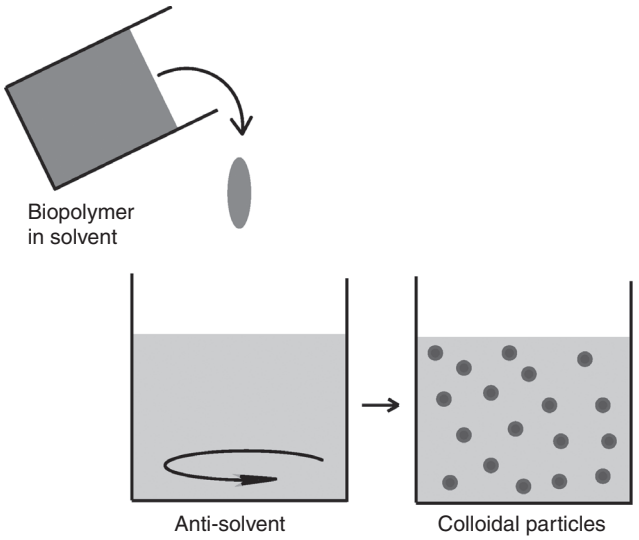

Figure 3 Schematic overview of an anti-solvent precipitation process

Anti-solvent precipitation

The anti-solvent precipitation technique, also known as nanoprecipitation, liquid–liquid dispersion, or solvent displacement technique, was originally applied to encapsulation and was patented by Fessi *et al.* [38,39]. The technique is straightforward, fast and easy to perform in a single-step procedure. Particle formation using this technique is instantaneous and requires two miscible solvents, in which ideally both the biopolymer and the encapsulate dissolve in the first (the solvent), but not in the second (the anti-solvent). When the biopolymer containing solvent diffuses

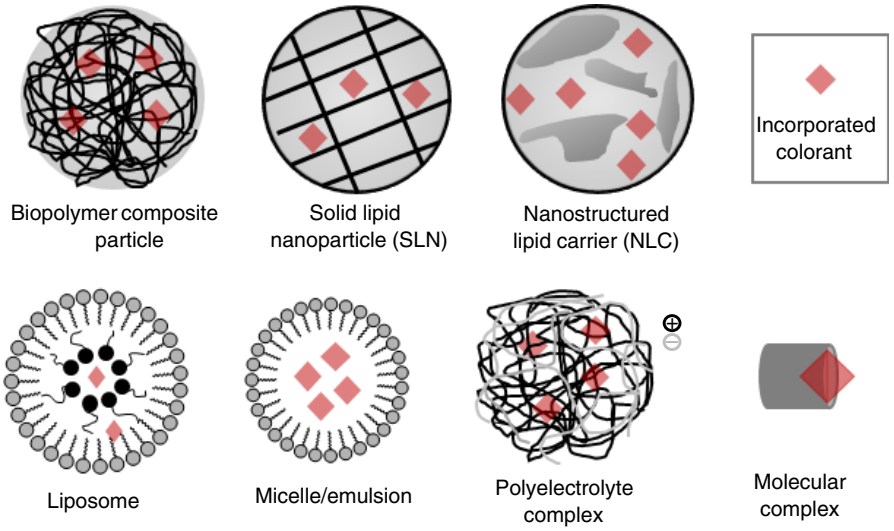

Figure 4 Examples of colloidal structures that are formed when an colorant is encapsulated using a biomaterial as encapsulant

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into the anti-solvent the biopolymer precipitates (Figure 3), which causes immediate entrapment of the encapsulate [40–42] (Figure 4). Particle sizes obtained following this procedure are often 20–300 nm depending on the specific materials and conditions used for the synthesis.

The term coacervation has also been used to describe this anti-solvent process. However, in the food industry and in colloid science this term is often used to refer to hydrophilic colloids which precipitate by interaction between two oppositely charged colloids upon chemical or physical triggers, such as manipulation of the temperature, salt, pH, and solubility conditions [43–46].

An advantage of the anti-solvent precipitation technique is that it does not require extended shearing or stirring rates, sonication, or very high temperatures, which is beneficial for biopolymers and sensitive encapsulates that could be affected by these conditions. Moreover, surfactants are not always necessary for stability, and toxic organic solvents are generally excluded from this procedure [42]. A possible disadvantage is that the technique is mostly suitable for compounds of a hydrophobic nature. However, when materials with these properties are used, this method often leads to very high entrapment efficiencies because of close to zero leakage due to the anti-solvent. For the application in dry form, particles synthesised using anti-solvent precipitation then need to be dried. This could be achieved by using freeze-drying or spray-drying as an extra step during particle production to keep particle deformation to a minimum. An example of this was in principle carried out by Patel *et al.* [23]. They prepared quercetin-loaded biopolymeric colloidal particles by precipitating quercetin (water-insoluble) and zein (hydrophobic protein) simultaneously by adding their hydro-alcoholic solution to the aqueous anti-solvent in the presence of sodium caseinate as a stabiliser.

Anti-solvent precipitation is commonly used to fabricate particles using a simple batch method on a laboratory scale; however, this is not directly suitable for industry, where continuous production is more desirable. Recently, a continuous technique based on anti-solvent precipitation was developed by Li *et al.* [47] for production of zein colloidal particles using flash nanoprecipitation with controlled particle sizes on a large scale. Later, Ebert *et al.* [48] also studied the potential of producing other protein particles by anti-solvent precipitation using a continuous dual-channel micro-fluidisation method.

Types of encapsulants suitable for food applications

Encapsulants that are suitable for food applications must be food grade, biodegradable, and stable in food systems during processing, storage and consumption. The most suitable encapsulants for food applications are carbohydrates, lipids, proteins, or mixtures thereof. As discussed previously, encapsulation can be achieved via several processes that lead, depending on the chosen technique and materials, to the formation of particles with various structures (Figure 4). These will be discussed next, along with examples of colorants that were successfully encapsulated.

Carbohydrates

In this section, different types of carbohydrates are discussed with regard to their potential for encapsulating colorants. Carbohydrates consist of monosaccharides, oligosaccharides and polysaccharides. They are highly suitable for encapsulation of colorants for food applications because they are already widely used as safe and inexpensive food ingredients. Carbohydrates are considered to be a suitable shell under high temperature processes due to their temperature stability in comparison with lipids or proteins, which might melt or become denatured when subjected to higher temperatures [49]. Examples of suitable carbohydrates include starch, pectin, guar gum, gum Arabic, xanthan gum, chitosan, cyclodextrin and maltodextrin. These will be discussed further in the following sections.

Dextrins

Dextrins are polysaccharides used as food additives (E1400). They are produced from starch by partial hydrolysis. Dextrins are freely soluble or dispersible in water and are slightly soluble to insoluble in anhydrous alcohol (for

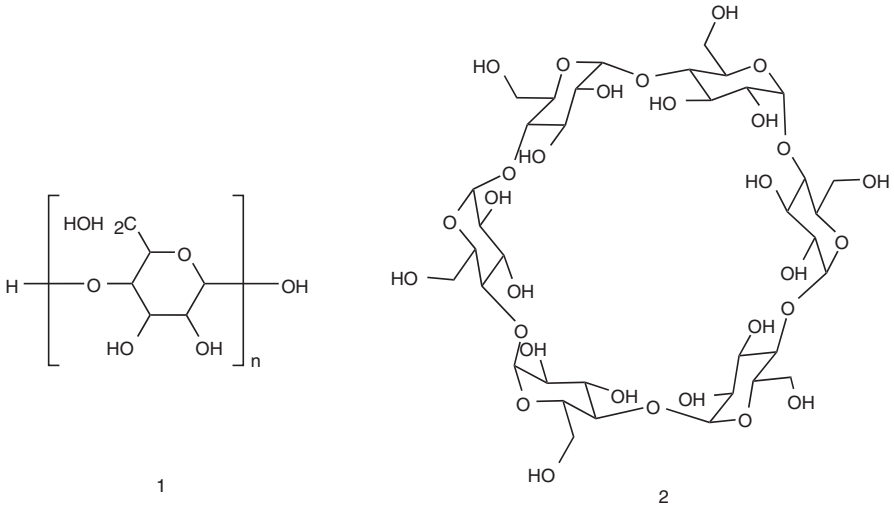

Figure 5 Chemical structures of (1) dextrin [50] and (2) α-cyclodextrin

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dextrin's molecular structure, see Figure 5.1) [50]. Maltodextrin also has the same structure as dextrin, with generally short molecular chains ($2 < n < 20$) [51]. Cyclodextrin has a cyclic chemical structure, of which the most abundant types are α-, β- and ϒ-cyclodextrin for six, seven and eight dextrin units, respectively (see Figure 5.2 for α-cyclodextrin). Classification of dextrins is by dextrose equivalent (DE); generally they have a DE between 3 and 20. Advantages of the use of dextrins as encapsulants are their low cost, blandness in flavour, and good flavour protection against oxidation. Hydrolysed starches are reported to improve the shelf life of orange oil and carrot carotene [52]. Otálora *et al.* [53] used spray-drying as an encapsulation technique for the encapsulation of betalains from cactus fruit using maltodextrin and cactus cladode mucilage as encapsulants. The stability of betalain is affected by several factors such as pH, water activity, and exposure to oxygen, light, temperature and degradative enzymes [54]. Microencapsulation had been proposed as a successful strategy to improve betalain stability, to make it easier to handle during processing for the use of food colourings. Using the spray-drying technique, particle sizes from 6–50 μm were obtained, and cactus cladode mucilage addition to maltodextrin yielded a more homogeneous particle size distribution. Particles that were produced with only maltodextrin were able to form a denser and more oxygen-permeable wall system, providing better storage stability for the betalain biological pigments compared with particles with a combination of maltodextrin and cactus cladode mucilage. Dextrin is often used in combinations with other encapsulants, as discussed in the following sections covering Gums and Starch.

Gums

Gum Arabic is a natural gum which is collected from the sap of various species of the acacia tree. It is a complex mixture of glycoproteins and polysaccharides and is soluble in water. Gum Arabic is edible, is used in the food industry as a stabiliser (E414), and is a well-known encapsulant which offers protection to encapsulated materials, as was reported by Krishnan *et al.* [55]. They spray-dried cardamom oleoresin using gum Arabic, maltodextrin and modified starch, and concluded that gum Arabic as an encapsulant increased the stability of the encapsulate the most. Gum Arabic is used as a steric stabiliser in many applications such as printing, paint production, glue and cosmetics. The key disadvantage of gum Arabic is its high cost and limited supply.

Guar gum (also called guaran) is a galactomannan polysaccharide (see Figure 6.1 for its chemical structure) which is extracted from guar beans; it is a natural product and it is edible (E412). Guar gum has thickening and stabilising properties which are useful in food, feed and industrial applications. Kuck and Noreña [56] studied the encapsulation of anthocyanins and phenolic compounds from Bordo grape skin by using different combinations of gum Arabic, partially hydrolysed guar gum and polydextrose as encapsulants. They obtained powders with retention of phenolic and anthocyanins greater than 80%. In addition, high antioxidant activities were also observed, representing a potential dye for use in functional foods. Considering the set of results, the spray-dried treatment with 5% partially hydrolysed guar gum and 5% polydextrose presented the best behaviour; this is due to the better retention of phenolic compounds, anthocyanins, and antioxidant activity.

Guar gum has similar properties to xanthan gum (Figure 6.2), which is also used in food applications (E415). Ravichandran *et al.* [57] studied the effect of different encapsulants on the stability and colour of freeze- or spray-dried betalains. Encapsulation of betalains, betacyanins and betaxanthins was performed using spray-drying, and the encapsulants used were combinations of maltodextrin, gum Arabic, pectin, and xanthan gum. The authors note that it was not possible to use guar gum as an encapsulant in spray-drying, possibly due to its high viscosity. Also, they found that encapsulation of betalains with maltodextrin, in combination with other gums, increases the stability of the betalains. Better colour properties were obtained by using maltodextrin, but the authors state that it is better to use mixtures because of colour stability.

Rajabi *et al.* [58] investigated the shelf-life improvement by encapsulating a saffron extract using maltodextrin, gum Arabic and gelatin as encapsulants. The main components in saffron are crocin, picocrocin and safranal, all of which degrade when exposed to light, heat and oxygen. An increase in the total solids content (from 30 to 40%) had a positive influence on the encapsulation efficiency and the saffron active component retention. The researchers also

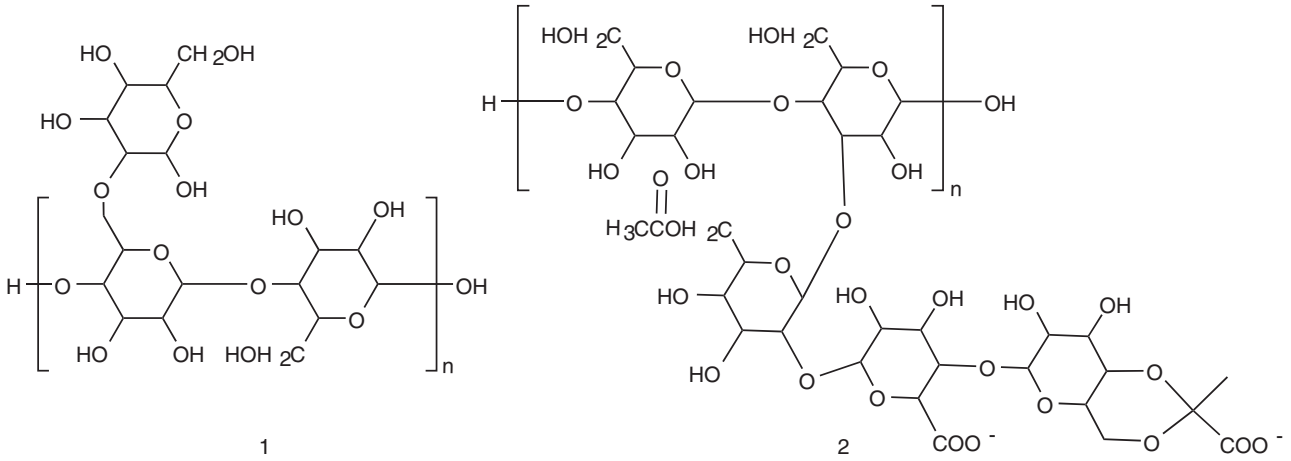

Figure 6 Chemical structures of (1) guar gum [60,61] and (2) xanthan gum [62,63]

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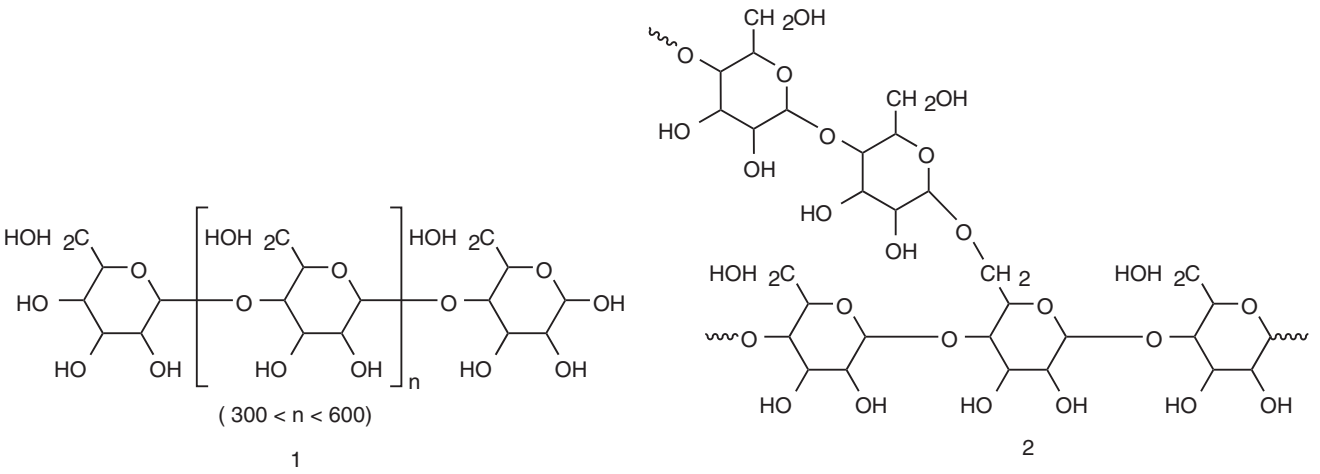

Figure 7 Chemical structures of (1) the linear amylose and (2) the branched amylopectin

synthesised particles with blends of the encapsulants, in which they found that the stability of the saffron active components increased as the quantity of gelatin decreased in the blend with maltodextrin and gum Arabic up to 1% (w/w). The final synthesis was optimised to contain 40% total solids, consisting of maltodextrin, gum Arabic and gelatin in the w/w ratio of 0.94:0.05:0.01. Akhavan Mahdavi *et al.* [59] studied the influence of different types of encapsulants (maltodextrin and gum Arabic, maltodextrin and gelatin, and maltodextrin) on the encapsulation of anthocyanins extracted from fresh barberry fruits (*Berberis vulgaris*). Among the encapsulants tested in this study, the combination of maltodextrin and gum Arabic led to a higher efficiency compared with the combination of maltodextrin and gelatin, or maltodextrin on its own; thus, it provided better protection of the anthocyanin biological pigments. The researchers concluded that all encapsulants demonstrated approximately the same physical properties.

Starch

Starch is a polymeric carbohydrate that can be fractionated in two main components, amylose (a linear chain) and amylopectin (a highly branched chain) [64], as shown in Figure 7, in which the specific composition differs between plants. Starch is produced by most green plants as energy storage. Farrag *et al.* [41] compared starch of different botanical origins (pea, corn and potato) as encapsulants for quercetin using anti-solvent precipitation. Starch origin has a significant effect on the behaviour of starch at nanometric level, as starches of different origins have different amylose/amylopectin ratios. It was found that the corn starch-quercetin nanoparticles had the lowest loading percentage of quercetin, the lowest antioxidant activity, and the lowest quercetin release kinetics. The nanoparticles of potato starch were very similar to pea starch nanoparticles, the only difference being that the potato starch nanoparticles showed a slightly higher loading capacity. Unfortunately, the researchers did not study the colour stability of the particles. However, a patent was filed in 1995 [65], in which quercetin-cyclodextrin complexes were formed with improved colour stability. Herein, spray-drying was used to encapsulate β-carotene with modified and unmodified tapioca starch, and maltodextrin by Loksuwan [52]. Powders obtained from modified tapioca starch demonstrated higher cold water solubility than unmodified starch. The total β-carotene content in the encapsulated core was highest for modified tapioca starch as an encapsulant, while it was lowest for maltodextrin as an encapsulant.

The stability of lycopene can be improved by spray-drying it with modified starch as encapsulant. Rocha *et al.* [66] found in their studies that although the encapsulation efficiency is relatively low (21–29%), the stability during storage of the lycopene improved significantly compared with free lycopene.

Starch is not only used on its own as an encapsulant, but also in combination with other carbohydrates. A comparative study was performed by Idham *et al.* [24], in which three different carbohydrates (maltodextrin, gum Arabic and soluble starch) were used for the encapsulation of anthocyanins, extracted from *Hibiscus sabdariffa*, using the spray-drying technique. The researchers found that a combination of maltodextrin and gum Arabic as encapsulants had the highest encapsulation efficiency, highest improved storage stability, and smallest change in the natural dye colour.

Lipids

Among the encapsulation systems, the lipid-based particles are promising systems for encapsulation and delivery of generally poorly soluble ingredients. For the application of encapsulation colorants using lipids, research is sparse to date. Moreover, particle or vehicle synthesis is often performed by a different method to the more conventional techniques (spray-drying or electrospinning; however, anti-solvent precipitation is possible). Therefore, lipid particles that offer potential use for the application of colorant encapsulation, namely, solid lipid nanoparticles, nanostructured lipid carriers and liposome vesicles, are discussed below.

Solid lipid nanoparticles

Solid lipid nanoparticles (SLNs) were developed as an alternative carrier system to the existing traditional carriers such as emulsions, liposomes and polymeric particles. SLNs are aqueous colloidal dispersions of which the matrix consists of solidified lipids that are highly structured [67] (Figure 4). Typical particle sizes of these SLNs are in the

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range of 40–1000 nm [68], which means that these particles can be microparticles or nanoparticles. The particles are prepared either with physiological lipids, or with lipids generally recognised as safe (GRAS). The primary advantage of SLNs is that they provide protection for sensitive molecules from the external environment (water, light). The disadvantages are their particle growth is not always controllable, their unpredictable gelation tendency, their unexpected dynamics of polymorphic transitions, and their inherent low incorporation rate due to the crystalline structure of the solid lipid [69,70].

In a study by Tiyaboonchai *et al.* [71], curcuminoid extract was successfully incorporated into SLNs via a microemulsion technique at moderate temperatures, achieving a mean particle diameter of 450 nm and an encapsulation efficiency of 70%. Furthermore, this study demonstrates that the stability of curcuminoids in a cream containing curcuminoids incorporated into SLNs was significantly improved compared with free curcuminoids in a similar cream formulation. The light and oxygen sensitivity of curcuminoids was strongly reduced by incorporating the curcuminoids into SLNs. Thus, SLNs are an attractive carrier system for light- and oxygen-sensitive substances.

More recently, Qian *et al.* [72] studied the physical and chemical stability of SLNs in comparison with liquid lipid nanoparticles, which both contained encapsulated β-carotene. Cocoa butter and hydrogenated palm oil were used as lipid encapsulants, and Tween 80 was used as a surfactant. The liquid lipid nanoparticles were stable, showing little extra particle growth during storage, whereas the particle size of SLNs increased due to aggregation. The rate of β-carotene degradation, or colour loss, in the liquid lipid nanoparticles was shown to be less than in the SLNs. Overall, these results show that the SLNs developed in this work did not provide any significant advantages over liquid lipid nanoparticles for the encapsulation and protection of β-carotene. A possible reason proposed by the authors is that the crystals formed by cocoa butter and hydrogenated palm oil mixtures were too highly organised, which led to the expulsion of β-carotene. Selection of alternative lipid mixtures which form a less highly organised solid phase may be able to overcome these problems, but further work is clearly needed to demonstrate this.

Nanostructured lipid carriers

A new generation of lipid nanoparticles has been developed as an improvement on SLNs [67], namely, nanostructured lipid carriers (NLCs). They are made of biocompatible and biodegradable lipids with well-established safety profiles and toxicological data [73,74]. NLCs consist of a blend of solid and liquid lipids that creates a less ordered matrix (see Figure 4), which allows a higher loading capacity and a lower possibility of drug expulsion during storage compared with SLNs [68]. The synthesis process can be modified to yield lipid particle dispersions with a 30–80% solid content [69].

Lacatusu *et al.* [75] produced NLCs using omega-3 fatty acid-rich fish oil to encapsulate lutein. Lutein is a natural colorant that is present in various vegetables. However, lutein is an unstable molecule with poor solubility in aqueous media. In this study, particles were successfully prepared via melting emulsification coupled with high shear homogenisation. The researchers concluded that the fish oil plays an important role in improving the antioxidant capacity, scavenging up to 98% of the oxygen-based free radical species. They also concluded that this represents a new and effective strategy for lutein delivery, which might be beneficial for the food sector. The formulation of lutein into colloidal nanolipid-in-water dispersions could serve to successfully incorporate this colorant into water-dispersible food systems.

Liposomes

Liposomes are closed vesicular structures consisting of bilayers of hydrated phospholipids. The bilayers are separated from one another by aqueous domains and they enclose an aqueous core. Liposomes' vesicles are able to entrap compounds of a different solubility, not only hydrophobic, but also hydrophilic molecules [76]. Hydrophobic ingredients will be entrapped in the double layer of the liposomes, whereas hydrophilic ingredients will be entrapped in the aqueous core of the liposome (see Figure 4). Liposomes can help to overcome the difficulty of the dispersion of hydrophobic substances in food formulations which are predominantly water-based. It is possible to entrap essential oils, hydrophobic peptides and carotenoids in liposomes. Additionally, the basic liposome structure of hydrated phospholipid bilayers can be modified with respect to the physical and chemical composition of the vesicle. This has resulted in extensive investigation into the use of liposomes for various applications in radiology, cosmetology and vaccinology. Particle sizes of liposomes for these applications typically range from 25 nm up to several μm and are usually dispersed in an aqueous medium [70,77].

In a recent study [77], β-carotene-loaded liposomes were prepared using proliposomes. The dispersions were stabilised using xanthan and guar gums as thickeners and the prepared vesicles had a diameter of 2000 nm. The β-carotene was protected from degradation by the liposomes for a period of 95 days and 90% of the encapsulated carotenoids were preserved after this time. The liposomes with carotenoids were incorporated in yogurt and the texture of this formulation showed no difference in viscosity to the reference yoghurt sample. Although sensory colour analysis shows that some optimisation must be performed regarding the observed colour of the final product, the researchers conclude that β-carotene-loaded liposomes can be a suitable option for producing the red colour in food products such as yoghurt.

Proteins

Proteins are ideal for use as encapsulants for microparticles and nanoparticles that have future use in food systems because they are GRAS and also have high nutritional value [78]. However, the nutritional value varies depending on the specific protein source [79]. Most proteins can be easily digested by the human gastrointestinal tract [80]. Protein microparticles and nanoparticles can be produced via many different techniques, including anti-solvent precipitation, pH-induced precipitation, spray-drying, electrospinning and emulsification. Particle sizes that can be obtained using proteins as encapsulants vary from 10 nm to 10 μm depending on the specific protein and particle synthesis

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technique which is used. Both animal and plant proteins have been used as encapsulants. The most commonly used animal proteins include gelatin, casein and whey protein. Plant-derived proteins commonly used for encapsulation purposes include soy protein, gliadin and zein [81].

Milk proteins

Milk proteins are biopolymers that are categorised into two main groups, caseins and whey proteins. Caseins [82,83] are proline-rich, open-structured proteins which have distinct hydrophobic and hydrophilic domains; 95% of the caseins are naturally self-assembled into casein micelles [84], spherical colloidal particles with a diameter of 50–500 nm. The major whey protein in cow milk, β-lactoglobulin, is a small globular protein [85]. It is folded up into an eight-stranded antiparallel β-barrel with a three-turn α-helix on its outer surface. Milk proteins are widely available, inexpensive, natural, and GRAS raw materials with high nutritional value and good sensory properties. They have many structural properties and functionalities which make them highly suitable as vehicles, or as components for the construction of vehicles, for delivering various bioactives [86]. Typically, milk proteins form molecular complexes with the ingredients they encapsulate (see Figure 4).

It has been reported [87] that curcumin was successfully encapsulated in microparticles based on pH-dependent solubility characteristics of curcumin and dissociation and re-association properties of sodium caseinate. The synthesised particles with a diameter of 200 nm displayed significantly enhanced dispersibility and bioactivity after the incorporation in casein micelles. Properties such as the solubility and photostability of several bioactive ingredients were reported to improve after binding with whey proteins [88]. The interactions between curcumin and β-lactoglobulin in aqueous solution have been studied and confirmed, and it was reported that this principle might also improve the antioxidant activity of curcumin [89].

A recent study [88] shows the encapsulation of curcumin in whey protein microparticles by spray-drying. This improved the solubility, and thereby the bioavailability, of curcumin. In solutions, curcumin formed soluble complexes with whey protein isolate via hydrophobic interactions, and its solubility increased linearly with whey protein isolate concentration. Solutions of these complexes were spray-dried as uniform microparticles and nearly 100% curcumin was retained in an amorphous state. Another study [90] showed that spray-drying of blueberry pomace extract (anthocyanins) in whey proteins improved the stability upon storage. The researchers measured a slight increase in antioxidant capacity and a two-fold increase in the total phenolic concentration upon storage.

Gelatin (mixed with various carbohydrates)

Gelatin is a mixture of peptides and proteins [91] produced by partial hydrolysis of collagen extracted from the skin, bones and connective tissues of animals. Gelatin readily dissolves in hot water and sets to a gel on cooling. When added directly to cold water it does not dissolve well. Gelatin and gum Arabic are often used together because of their opposite charges at low pH, which causes attraction and thereby, by also lowering the temperature, the formation of insoluble particles [92]. Hydrophilic substances such as gelatin could also be used to encapsulate hydrophobic substances, including vegetable oils or oil-soluble colorants. Anti-solvent precipitation of gelatin could be performed in ethanol instead of water to form particles. Encapsulation of colorants in gelatin is often carried out by spray-drying and used in combination with carbohydrates, as was described earlier in the section on Gums. An example is from Shu *et al.* [93], who spray-dried lycopene using gelatin and sucrose as encapsulants and found an increased storage stability for lycopene. Another option is using an emulsification technique, as described by Medeiros *et al.* [94]. They used emulsification to encapsulate a carotenoid extract from Cantaloupe melon pulp in gelatin. This was proposed as a strategy to improve the solubility of carotenoids in water. They applied the particles to a yogurt sample and found improved colour stability compared with the colour effects of the crude extract.

Soy proteins

Soy proteins are hydrophilic proteins that supply desirable functional properties when added to a variety of foods, for example, emulsification, fat absorption, moisture holding, thickening and foaming. Soy protein isolate is negatively charged in solutions above its isoelectric point (pH~4.5) [95–97]. A recent study [98] has shown the encapsulation of capsanthin by soy protein isolate and chitosan through complex coacervation. Capsanthin possesses excellent colouring performance and health benefits; however, its application in the food industry is limited due to its susceptibility to humidity, heat and light. The researchers showed that capsanthin was successfully encapsulated by soy protein isolate: chitosan coacervation and the encapsulated capsanthin displayed enhanced stability against low and medium moisture, heat, and light. In a spray-drying study [99], soy protein isolate proved to have the highest encapsulation efficiency and the best stabilisation of phytochemicals extracted from wild blueberry pomace among the protein-rich sources tested (the others were wheat flour, chickpea flour and coconut flour). The researchers link the high encapsulation efficiency to the enhanced capacity of soy protein isolate to bind and complex the blueberry polyphenols. They even state that in addition to binding and complexing with the blueberry pomace extract polyphenols, the soy protein isolate efficiently migrates to the surface of polyphenol-protein particles, thereby acting as an efficient drying carrier and eliminating the need for popular polysaccharide-based drying carriers such as maltodextrin or chitosan.

Gliadin

Gliadin is a plant protein that has been widely used for particle production and encapsulation using the anti-solvent technique. It is a proline-rich storage protein from wheat, and is therefore sometimes also called wheat gluten [100,101]. It is insoluble in water but is soluble in alcohol-water mixtures. A potential advantage of utilising water-insoluble proteins is that no additional hardening step is required to preserve their integrity in aqueous-based products. However, a potential drawback of using gliadin is that a part of the population has gluten intolerance, and therefore some people have been promoting the elimination of gluten proteins from their diet completely [102].

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Davidov-Pardo *et al.* [103] encapsulated resveratrol using gliadin in combination with the hydrophilic pectin by utilising the simple anti-solvent precipitation technique. Resveratrol is a natural polyphenol that is extracted from the skin of grapes, blueberries, raspberries and mulberries. While resveratrol is not a colorant, it is used as a dietary supplement, and this research demonstrated the use of gliadin as an encapsulant for food applications. The use of the hydrophilic biopolymers improved the encapsulation efficiency. The authors concluded that these particles appear to be promising encapsulation systems for enriching functional foods with this nutraceutical compound. In a second study by the same researchers [104], the influence of the encapsulation on the degradation of resveratrol by ultraviolet (UV)-light was investigated. It was concluded that all particle formulations protected resveratrol against UV-light to some extent, which is also a very promising result for light-sensitive colorants. Another group encapsulated curcumin with gliadin using lecithin as a stabiliser [105]. They managed to reach a high encapsulation efficiency (77–90%) and improved stability against UV-light and temperature. Researchers in another recent study [106], instead of using a simple anti-solvent technique, utilised a pH-cycle technique to encapsulate β-carotene in gliadin using xanthan gum as a stabiliser. The presence of the xanthan gum was critical for the improvement of the stability against creaming. Xanthan gum also had a positive effect on the chemical degradation during storage.

Zein

Zein [107] is the major storage protein from corn. It belongs to the class of prolamins (that also includes gliadin, hordein and kafirin) [108,109]. Zein is insoluble in pure water or in pure ethanol; however, it is soluble in an ethanol-water mixture [110,111]. Zein can easily be converted into spherical microparticles and nanoparticles via a simple anti-solvent technique or electrospinning [112–114]. Because of its good barrier properties, biodegradability, and biocompatibility, zein has been extensively researched for encapsulation of various substances (including drugs, essential oils, colorants and enzymes) [115,116]. The simplicity of generating zein colloidal particles (often via anti-solvent precipitation), coupled with the ease of loading with functional ingredients, make it an attractive encapsulant. This has resulted in zein having recently received widespread interest for the generation of functional colloidal structures [117].

In a recent study [118], the possibility of using white zein particles as a replacement for titanium dioxide in liquid food systems was investigated. It was possible to purify zein protein and synthesise particles via anti-solvent precipitation to obtain stable white colloidal particles. Their scattering properties could be tuned by varying the particle size and concentration such that these particles could be used as clouding agents replacing titanium dioxide, albeit at a higher concentration. As discussed earlier in this review, the safety of titanium dioxide is under discussion and consumers prefer alternatives to this inorganic colorant. An example of a food product which contains titanium dioxide is mayonnaise. It was reported in the Dutch National Food Consumption Survey (DNFCS) that the oral intake of titanium dioxide originates for a relatively large part (7% in the age group 7–69 years old) from mayonnaise [12]. Gu *et al.* [119] proposed that zein particles could be used as a fat replacement in mayonnaise. They based their conclusion on appearance, stability and total colorimetric values, as well as on rheological, microstructure, and sensory analysis. This implies that purified zein particles could also be suited for use as a replacement for titanium dioxide in mayonnaise.

Patel *et al.* [120] showed that it was possible to create a tunable colloidal colorant which ranged from yellow, to green, to blue. They encapsulated curcumin and indigo-carmine into zein protein particles using anti-solvent precipitation. The different colour shades were obtained by loading different ratios of curcumin and indigo-carmine inside the particles. More importantly, the entrapment of these colorants in colloidal particles resulted in the enhancement of the stability of both curcumin and indigo-carmine to photodegradation. Another study [121] demonstrated that it was possible to synthesise zein particles loaded with lutein and stabilised by lecithin and Pluronic F 127 surfactants via anti-solvent precipitation. The addition of surfactants increased the particle size and improved the polydispersity index. The zein particles were able to protect lutein from degradation under various storage conditions, as compared with the emulsified lutein. This complex formulation provided good protection against direct UV-light exposure for at least 10 h. Wang *et al.* [122] used anti-solvent precipitation to encapsulate β-carotene with zein, carboxymethyl chitosan and tea polyphenols. They tested the solubility of the freeze-dried powders in water, which is one of the factors used to evaluate their practical application for the food industry. Redispersibility was measured by absorption in time after adding the freeze-dried particles to water. It was found that the combination of β-carotene with zein, carboxymethyl chitosan and tea polyphenols was the best combination of encapsulants compared with β-carotene with the combination of zein and carboxymethyl chitosan without tea polyphenols, or β-carotene with zein alone. These results show that it is possible to use particles of zein, carboxymethyl chitosan and tea polyphenols as a potential carrier in the food industry.

Conclusions and Outlook

The various research results discussed in this review show that colorant encapsulation can be achieved by using different methods, of which we have discussed three main techniques, spray-drying, electrospinning and anti-solvent precipitation. Spray-drying is used to obtain dry microparticles and nanoparticles (but mainly microparticles), electrospinning for dry microparticles and nanoparticles, and anti-solvent precipitation for microparticles and nanoparticles dispersed in liquid, which can also be dried by using either spray-drying or freeze-drying. They are all techniques that are already used in industry or which have possibilities for scaling up. The biopolymers that are most suitable for use as encapsulants for the three discussed encapsulation techniques are mainly carbohydrates and proteins.

In this review, examples of successful encapsulation of natural colorants using carbohydrates, lipids and proteins as encapsulants were discussed. The research results described reveal that encapsulation of natural colorants

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using biopolymers provides significant protection for these colorants against environmental conditions such as increased temperatures, oxidation and photodegradation. Also, stability can even be further improved by co-encapsulating antioxidants and other stabilisers to the particles.

The scalability of the discussed encapsulation techniques demonstrates the potential of industrial scale production of fully natural encapsulated particles. They can satisfy consumers' desires for natural ingredients, i.e. the natural origins of both encapsulants and colorants. Moreover, their improved stability upon encapsulation facilitates future implementation of these encapsulated colorants in consumer products, not just in the food industry, but also in the pharmaceutical, cosmetic, textile, and other industries.

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