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Review

Microencapsulation of Anthocyanins—Critical Review of Techniques and Wall Materials

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Abstract: Anthocyanins are value-added food ingredients that have health-promoting impacts and biological functionalities. Nevertheless, there are technological barriers to their application in the food industry, mainly because of their poor stability and susceptibility to harsh environmental conditions, such as oxygen, temperature, pH, and light, which could profoundly influence the final food product′s physicochemical properties. Microencapsulation technology is extensively investigated to enhance stability, bioaccessibility, and impart controlled release properties. There are many varieties of microencapsulation methods and diverse types of wall materials. However, choosing a proper approach involves considering the processing parameters, equipment availability, and application purposes. The present review thoroughly scrutinizes anthocyanins′ chemical structure, principles, benefits, and drawbacks of different microencapsulation methods, including spray drying, freeze drying, electrospinning/electrospraying, inclusion complexes, emulsification, liposomal systems, ionic gelation, and coacervation. Furthermore, wall materials applied in different techniques plus parameters that affect the powders′ encapsulation efficiency and physicochemical properties are discussed. Future studies should focus on various processing parameters and the combination of different techniques and applications regarding microencapsulated anthocyanins in functional foods to assess their stability, efficiency, and commercialization potentials.

Keywords: anthocyanins; microencapsulation methods; encapsulation efficiency; biopolymers; wall materials

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1. Introduction

The far-reaching implications of synthetic colorants on human health are the prime motivation for the rising interest in plant-derived compounds, highlighting the need to seek suitable alternatives for the pharmaceutical and food sectors [1].

Anthocyanins are nontoxic pigments, soluble in aqueous food systems, and belong to the flavonoid class. Apart from imparting food color, anthocyanins possess a broad array of biological activities and therapeutic effects, such as antimicrobial activity, antioxidant protection, anti-inflammatory, antidiabetic, antiobesity, and anticarcinogenic impacts [2]. These phenolic compounds are responsible for brilliant colors ranging from pink, red, and orange to blue in flowers, seeds, fruits, and peels of numerous plants [3]. Anthocyanins possess a C6–C3–C6 basic skeleton (flavylium cation) and consist of an anthocyanidin (aglycone unit) linked to sugar, typically at the 3-position on the C-ring and methoxyl and hydroxyl groups [4]. Up to now, 23 types of anthocyanidins have been recognized and their structures can differ in terms of the substitutions at positions 5′ and 3′. The most widespread and commonly occurring aglycon forms (anthocyanidins) in fruits and vegetables are cyanidin, delphinidin, pelargonidin, peonidin, malvidin, and petunidin (Figure 1), with distributions of 50%, 12%, 12%, 12%, 7%, and 7%, respectively [2]. The solubility of anthocyanins in water, ethanol, and to some extent, in methanol depends on the presence of sugar(s) and hydroxyl groups on the rings [5]. The most prevalent sugar linked to anthocyanidins is glucose, while arabinose, galactose, rhamnose, and rutinose have

also been detected. These sugars could also be acylated with various aliphatic/aromatic acids [6]. The number, type, and position of the attached sugar; the position and number of methoxyl and hydroxyl groups; the number, nature, and extent of organic acids linked to the sugar lead to the various structure of the anthocyanin molecules [7]. The incorporation of anthocyanins into food systems yet faces technological obstacles in the food industry, as these promising molecules are unstable and highly likely to degrade in the presence of light and oxygen, high temperatures between 100 and 121 °C, and slightly acidic to neutral conditions, resulting in poor bioavailability and color fading during processing and storage [5,8,9].

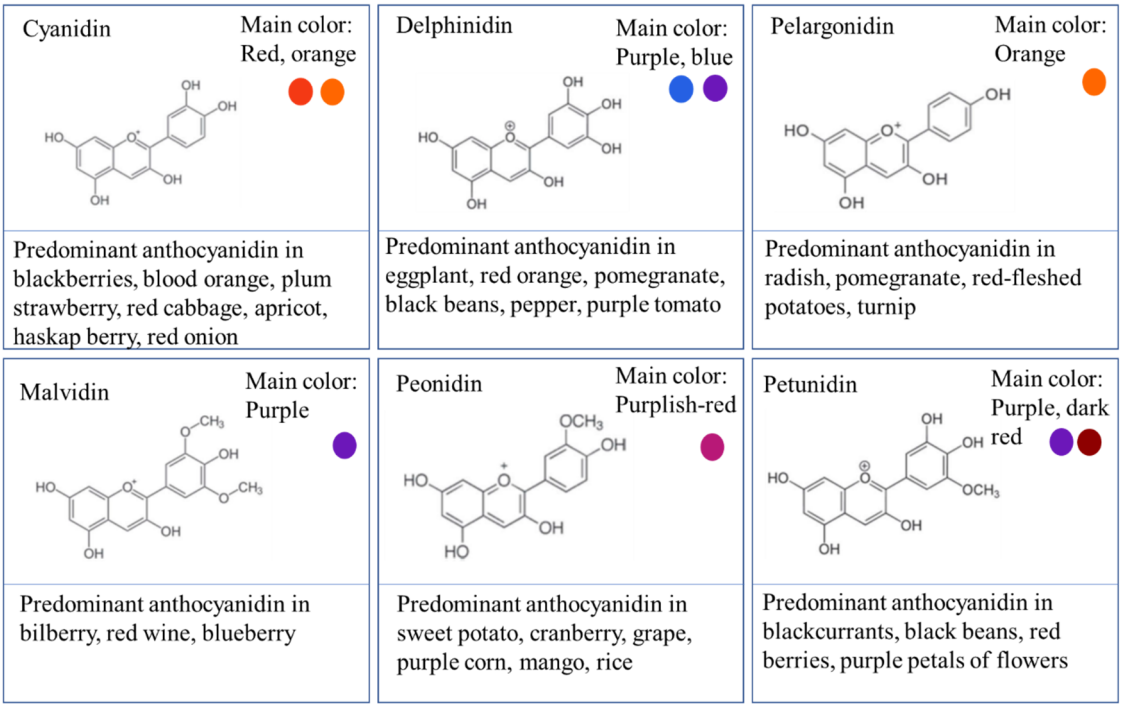

Figure 1. Chemical structures, colors, and sources of the most important anthocyanidins [2].

Microencapsulation is a satisfactory resolution regarding effectively protecting anthocyanins against adverse environmental conditions, and at the same time, providing physical stability, controlled release over time, enhanced bioaccessibility, and ease of handling and storage. In this process, a multicomponent structure is formed via entrapment of the solid, liquid, or gaseous substances as an active core material into a polymer as a wall or carrier material [10–12]. The products achieved using this process are different in size, ranging from 1 to 1000 µm. The morphologies generated via microencapsulation are also diverse, including microspheres, microcapsules, and microparticles (Figure 2). In microspheres, the core material is dispersed in the matrix and on the surface, which is different from microcapsules. Microcapsules could either be mono- or polynuclear, where in the former, a single active compound is coated by a wall material, while the latter consists of two or more cores. The wall material with which the active ingredient is coated could be mono- or multilayered. Concerning microparticles, they might have an irregular shape [13].

Up to now, multifarious strategies have been successfully investigated for the microencapsulation of anthocyanins (Table 1), where the most widely employed methods are as follows:

1. Physical methods, such as spray drying, lyophilization (freeze drying), and electrospray/spinning.
2. Chemical methods, such as the formation of inclusion complexes.

3. Physicochemical approaches consisting of liposomes, emulsification, coacervation, and ionic gelation.

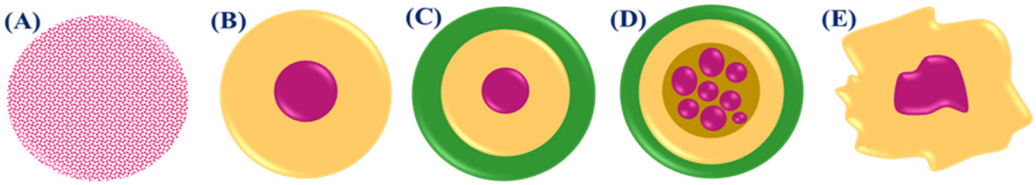

Figure 2. Schematic illustration of various morphologies formed by microencapsulation: microsphere (**A**), monolayer and mononuclear microcapsule (**B**), multilayer and mononuclear microcapsule (**C**), multilayer and multinuclear microcapsule (**D**), and microparticle (**E**).

Generally, the characteristics and interaction between core and wall materials, expected particle size, release properties, industrial scalability, and cost of the process are contributing factors in the proper selection of the technique. A wide range of biopolymers, such as proteins, polysaccharides, and lipids, have been utilized as wall materials of anthocyanins, irrespective of the microencapsulation techniques [14]. This review paper discusses the relative merits and provides information on the latest developments of the methods used for the microencapsulation of anthocyanins and a critical review of wall materials and influential parameters on encapsulation efficiency and physicochemical properties.

Table 1. Overview of the advantages, disadvantages, and variable parameters of various microencapsulation methods.

Method	Benefits	Drawbacks	Variable Parameters	Reference
Physical Methods				
Spray drying	Rapid, versatile, cost-effective, easy to scale up. High encapsulation efficiency, anthocyanin retention, and relatively good storage stability.	Production of nonuniform particles with a wide size distribution. Anthocyanin degradation and loss of product.	Operational parameters: feed flux, inlet and outlet air temperature, flow rate. Material properties: the type, concentration, and viscosity of wall material. Core to wall material ratio.	[15]
Freeze drying	Uses a low temperature.	Long processing time and high cost and energy. Low stability and sensitivity to oxidation.	Drying time and pressure.	[16]
Electrospraying/electrospinning	Cost-effective, one-step method. Suitable for heat-sensitive compounds. Produces particles with a high-surface-to-volume ratio, controlled release, improved functionality, and physical properties.	Slow, time-consuming, and produces low encapsulation yield. Hardly repeatable, especially at the industrial level.	Operational parameters: flow rate, electrical potential applied, and distance between the tip of the syringe nozzle and the collector. Intrinsic properties: surface tension, viscosity, concentration, molecular weight of the polymer, and electrical conductivity. Ambient conditions: humidity and temperature.	[17]
Chemical Methods				

Table 1. *Cont.*

Method	Benefits	Drawbacks	Variable Parameters	Reference
Inclusion complex	Preserves anthocyanins from polymerization, oxidation, and degradation reactions. Provides thermal stability and controlled release properties.	Low solubility of β-CD in water leads to precipitation or a viscous solution at high concentrations.	Inclusion temperature, solvent, structure, and polarity of the guest molecules and host cavity.	[10]
Physicochemical Methods				
Pickering emulsions	Ecofriendly and no surfactants are needed. Higher coalescence stability, strong protection, storage stability, and tailored release.	Industrialization.	Particle wettability. The oil phase and oil-to-aqueous phase ratio. Particle concentration and surface coverage. Particle charge, salt concentration, and pH. Emulsion preparation method, and temperature.	[18]
Liposome system	Increased adsorption and bioavailability. Nontoxic and nonimmunogenic.	Low encapsulation efficiency. Lipid oxidation, poor physicochemical stability, and wide particle size distribution. Postprocess step is required.	Source and concentration of phospholipid. Buffer pH.	[6,10]
Ionic gelation	Low cost and does not require advanced equipment, high temperatures, and organic solvents.	Rapid release when hydrophilic or low-molecular-weight materials are encapsulated.	Concentration of the crosslinking polyelectrolyte and wall material solution. Reaction and stirring time. Core-to-wall material ratio.	[19,20]
Coacervation	High loading capacity, low temperature operating requirements, reduced evaporation losses of volatile compounds, and thermal degradation. Tailored release of active compounds.	Produced particles are very sensitive to pH and ionic strength. Agglomeration of particles and particle size control. Complexity of the method.	Processing parameters: emulsion preparation method, temperature of the medium, pH and ionic strength, crosslinking and drying methods. Material properties: chemical composition, concentration, charge densities, molecular weights of polymers, and weight ratio.	[21]

2. Microencapsulation Techniques

2.1. Physical Methods

2.1.1. Spray Drying

Spray drying (SD) is one of the oldest and industrially common microencapsulation methods, and about 80 to 90% of anthocyanins have successfully been encapsulated via this technique [22]. SD consists of three key stages [23]:

1. Atomization of the fluid feed in the form of suspension, solution, or emulsion through a high-pressure nozzle into a drying chamber.
2. Direct contact between the sprayed droplets and the heating medium typically occurs at 150–220 °C, leading to evaporation of the solvent.

3. Separation and recovery of the powdered product from the air using a filter or cyclone.

This technique is rapid, versatile, cost-effective, easy to scale up, and generates high anthocyanin retention and relatively good storage stability. On the other hand, the atomization process produces particles with a high surface-to-volume ratio, which provokes thermal exposure and, consequently, degradation of anthocyanins and a reduction in the antioxidant activity of the microcapsules. Furthermore, nonuniform particles with a wide range of size distribution are produced, making the control over the morphology and size a demanding matter [10]. The particle size could generally be influenced by the spray atomizing pressure, nozzle position and size, liquid flow rate, polymeric structure, and solution concentration. The properties of the powder largely depend on numerous factors, such as the drying parameters, including the feed flux, air temperature, flow rate, and the type of wall material, as well as its concentration and viscosity [24].

The loading capacity of microcapsules is estimated using the encapsulation efficiency (EE). Temperature is the main factor that can affect the EE in this technique. High temperatures lead to a decline in the amount of powder that is adhered to the chamber wall, enhancing the EE. Furthermore, high temperatures facilitate the drying rate and formation of a crust, restricting the diffusion of heat in the droplet and the leaching of anthocyanins [25]. Souza et al. [26] concluded that high air temperatures, combined with a high solid concentration of the solution, results in a high EE and preservation of the anthocyanins. In a study by Laokuldilok et al. [27], *Oryza sativa* L. anthocyanins were microencapsulated and sprayed at 140, 160, and 180 °C. According to the results, as the inlet air temperature rose, the process yield increased from approximately 64% at 140 °C to 82% at 180 °C. Still, the total anthocyanin content (TAC), surface anthocyanin content (SAC), and radical scavenging capacity of the microcapsules decreased. It is worth pointing out that both the outlet and inlet temperatures are interrelated and should be considered while using SD because anthocyanins are highly heat-sensitive compounds [28]. A high inlet temperature exposes particles to an increased outlet temperature. In contrast, reduced outlet temperatures extend the drying time, creating a combined effect of temperature and time that directly impacts the loss of anthocyanins [29].

The EE of anthocyanins created using spray drying can also be affected by the core-to-wall mass ratio. Several studies have adopted a typical core-to-wall ratio of around 25% for obtaining the high EEs [30–33]. In contrast, Ortiz-Basurto et al. [34] reported that the core-to-wall ratio of 1:6 resulted in a higher yield (52.90–60.66%) in comparison to the ratio of 1:4 (25.40–43.14%) and 1:5 (35.40–52.08%). This finding might be associated with greater proportions of core materials close to the drying surface, decreasing the air–particle interface's diffusion path length [15].

The solid concentration of the wall material can also affect the EE of anthocyanins. On the one hand, it has been stated that as the solid concentration goes up, the viscosity of the wall material increases, where this phenomenon influences the atomization process and causes the production of large and elongated particles, affecting the drying rate [23]. Moreover, the formation of cracks on the particles' surfaces eases anthocyanins' diffusion from the core to the wall. Das et al. [25] utilized modified rice starch with concentrations between 5 and 10% (w/v) for the microencapsulation of anthocyanins of purple rice bran. It was reported that the EE dramatically went down once the concentration exceeded 7%. On the other hand, García-Tejeda et al. [35], who produced phosphorylated starch loaded with purple maize anthocyanins, reported the highest EE (49.11%) when the concentration was 20%. At the same time, this figure was lower for solutions with 10 or 15% concentrations.

The choice of proper wall material for microencapsulation via spray drying fulfills a prominent role in microcapsules' EEs. The selection of encapsulating agent in SD is somewhat limited because it should meet the following criteria: acceptable solubility, low viscosity at high concentrations, emulsifying properties, and film-forming capacity [23]. Wall materials that meet the requirements and are thus suitable for spray drying microencapsulation include:

- Polysaccharides (starch, chitosan, pectin, natural gums, mucilages, cellulose, and their derivatives).
- Animal and plant-based proteins (whey protein, caseinate, gelatin, and soy protein).

Generally, the contribution of polysaccharides is larger than that from proteins for microencapsulation purposes using spray drying. The reasons why polysaccharides are extensively considered as suitable wall materials are their high holding capacity, low viscosity, excellent solubility, and emulsifying properties [6].

Maltodextrin has been widely used as a wall material for the microencapsulation of anthocyanins using SD. In this direction, dextrose equivalent (DE) has a pivotal role in the process yield of spray drying since a higher degree of DE produces a large proportion of powders with a wetted surface and a high wall material thickness that sticks to the chamber wall and diminishes the process yield [27]. Furthermore, anthocyanins become more heat-sensitive while applying higher DE due to the increased number of open sides of the molecule, facilitating the oxidation of aldehydes and ultimately the production of a deformed structure [36]. Nayak and Rastogi [37] developed microcapsules with maltodextrin with 6, 19, 21, and 33 DE containing anthocyanins from Garcinia Indica. It was revealed that the anthocyanin content increased from 325 mg/100g with a DE of 6 to 430 mg/100 g with a DE of 21; nevertheless, both parameters dropped as DE rose from 21 to 33. Apart from DE, the concentration of maltodextrin can also influence the EE. Da Rosa et al. [29] examined the effects of maltodextrin's concentration on the properties of microcapsules containing blueberry anthocyanins. They found EE percentages ranging from roughly 74 to 85%, with the best EE reported for the highest concentration of maltodextrin (18%) at all temperatures.

In most of the studies, the highest EE and stability were achieved by using a combination of wall materials [31–33,38]. For instance, Da Rosa et al. [39] applied a standard formulation of maltodextrin and hi-maize starch combined with gum Arabic, inulin, and both gum Arabic and inulin for the microencapsulation of anthocyanins from a blueberry extract. The coupling of wall materials also led to considerably enhanced EE and stability. The lowest EE of 96.80% was recorded for the mixture of maltodextrin/hi-maize starch/inulin, while the superior EE, 98.83%, was observed for maltodextrin/hi-maize starch/inulin/ gum Arabic. The latter sample also had the lowest anthocyanin loss (4.45%, 5.82%, and 5.94% at freezing temperature, during refrigeration, and at RT, respectively) and the most prolonged half-life (913.41 days at freezing temperature, 693.32 days during refrigeration, and 679.81 days at RT) throughout the storage period. According to Silva et al. [40], 30% maltodextrin as a carrier agent had the best performance regarding anthocyanin retention compared to 25% Capsul™ + 5% maltodextrin and 25% Arabic gum + 5% maltodextrin. Rocha et al. [41] also used maltodextrin, gum Arabic, and whey proteins for the microencapsulation of blueberry, jussara, and jabuticaba anthocyanins. It was observed that the individual wall materials had EEs above 90%, with the highest belonging to maltodextrin, followed by whey protein concentrate and gum Arabic [41]. Kurek and Sobierskala [42] also reported the greatest EE of 94% for the lowest quantity of β-glucan (0.5%) as a carrier material for elderberry anthocyanins. In contrast, this figure for the combination of maltodextrin and Arabic gum was 80%. Hence, the choice of wall material for spray drying microencapsulation has an impact on the EE because of the disparity between carriers' properties in terms of viscosity, solid content, film-forming capacity, and cost.

Regardless of the benefits presented by this method, the high temperatures needed in the process can degrade the anthocyanins. Moreover, loss of product in the drying chamber wall can result in a significant waste of material. Furthermore, the nonuniform size distribution, the large particle size, and various morphological properties of powders restrict their food applications.

2.1.2. Freeze Drying

Freeze drying (FD), unlike SD, applies low temperatures, making it a potential candidate for the dehydration of water-soluble and thermosensitive bioactive compounds, particularly anthocyanins. The FD method's fundamental principle is sublimation, during which the moisture is directly converted from a liquid to a gas phase, and finally, a powder is formed [43]. Since this process is performed in a vacuum and at a low operating temperature, the number of deterioration reactions is dramatically reduced. Regarding this method's drawbacks, it is a time-consuming process (between 24–48 h) and requires a high amount of energy [44]. The major limitation of this approach is the production of a porous structure due to ice sublimation, paving the way for anthocyanins to easily contact oxygen through pores on the particles' surface [10]. Many studies have made a comparison between FD and SD microencapsulation of anthocyanins. Most of them have indicated no significant difference in the total anthocyanin content of microcapsules produced using both methods [45–47]. In FD, anthocyanins are mainly degraded due to grinding and oxidation processes. In contrast, in SD, the loss of anthocyanins is attributed to the high temperature, oxidation, and ultimately, polymerization and degradation [46]. However, some studies detected notably higher anthocyanin content in freeze-dried samples when compared to SD. In this regard, Laokuldilok and Kanha [27] found an approximately 72% greater quantity of black rice anthocyanins in microcapsules produced using FD. Furthermore, the process yield varied from 64.07 to 82.16% for SD but above 85% for FD. Murali et al. [48] also reported a 22–25% higher anthocyanin content for freeze-dried microcapsules loaded with black carrot juice, with the EE ranging from 80.81 to 98.51% for FD and 60.04 to 82.48% for SD. Yu and Lv [49], who attempted to microencapsulate rose residue anthocyanins, showed that the anthocyanin and total phenolic compound retention were, respectively, 91.44% and 95.12% for FD while 75.85% and 86.00% for SD.

The morphological properties (size and shape) of powders are the most noticeable difference between the FD and SD microcapsules. According to the survey by Laokuldilok and Kanha [50], FD resulted in irregular agglomerates and a thin porous sheet-like material. SEM observations of spray-dried particles also showed that high inlet temperatures led to small-sized spherical particle formation with a smooth surface, whereas low temperatures generated shriveled ones. Considering these properties, freeze-dried microcapsule had 19–100% lower storage stability than spray-dried samples at all temperatures due to their morphology and oxygen permeability [50]. In contrast, according to Fredes et al. [45], no difference was observed in the half-life of microencapsulated maqui juice through FD and SD despite the different morphological properties. The stability of powder during storage could also be evaluated using the T_g value. This parameter can be influenced by the moisture content, chemical structure, and molecular weight of the carrier agents. Hence, the most stable powder would have the highest T_g value. Therefore, to ensure stability, the powder's temperature should be lower than T_g. In this case, molecular mobility is limited since the material remains in the glassy state, which controls the chemical, physical, and biological changes [43]. Kuck and Noreña [46] reported T_g values ranging from 11 to 26 °C for grape skin extract microencapsulated via FD, whereas this figure for SD was between 42 and 52 °C. Thus, freeze-dried samples had lower stability, mainly due to the high moisture content.

The anthocyanin-to-polymer wall material ratio in lyophilization has a role to play in the EE and anthocyanin content of microcapsules. Mazuco et al. [51] revealed that the anthocyanin content and EE were 116.89 mg/100 g and 20.45%, respectively, with an anthocyanin-to-polymer wall material proportion of 2:1, followed by an increase to 151.68 mg/100 g and 21.11% in the ratio of 2:3.

The EE, stability, and physical properties of the powder can be profoundly impacted by the composition and type of wall material. Maltodextrin is the most popular wall material that has been widely used in this technique. Mixing this wall material with other encapsulating agents has been examined to effectively preserve anthocyanins and produce better physical properties. To illustrate, anthocyanins from jaboticaba pomace

were microencapsulated with a different mixture of wall materials, namely maltodextrin (MD), MD–pectin (PEC), MD–soy protein isolate (SPI), and MD–pectin–SPI [52]. The most desirable physical properties in terms of the lowest hygroscopicity and solubility were recorded for maltodextrin–pectin–SPI, followed by maltodextrin–soy protein isolate and maltodextrin–pectin. This observation was connected to the interaction of different wall materials with anthocyanins during the process. On the one hand, amine and carbonyl groups of proteins form hydrogen bonds with the anthocyanins' hydrophilic region. On the other hand, the hydrophobic region of proteins interacts with the benzene ring of the anthocyanin. In water, microcapsules' solubility diminishes because the nonpolar parts of anthocyanins and proteins interact via van der Waals forces, reducing the nonpolar region exposed to water. Furthermore, the wall materials protect the adverse effect of UV on the total monomeric anthocyanins and antioxidant capacity, with the best performance being reported for MD–PEC–SPI (up to 99%). In the study of Cai et al. [53], the combination of carboxymethyl starch (CMS) and xanthan (XG) gum resulted in an EE of over 96%. Still, there was no considerable impact when incorporating various xanthan gum ratios. It was concluded that if the wall material's viscosity surpassed the optimal value, the change in viscosity could not notably enhance the EE. The highest DPPH scavenging activity during incubation at 50 °C was reported for CMS/XG (30/1), followed by CMS/XG (60/1) and CMS/XG (90/1). Furthermore, the highest anthocyanin retention was observed for CMS/XG (150/1), which was 90.47% at 5 °C and 76.11% at 37 °C after 30 days. In the study of Mansour et al. [54], red raspberry anthocyanins were microencapsulated in SPI and gum Arabic. It was highlighted that using two wall materials was more effective than utilizing a single wall material in terms of the EE and stability. In this direction, blending 2.5% (w/v) of SPI and 2.5% (w/v) of gum Arabic led to the highest efficiency (98.87%) and thermal stability. Using a pure extract produced anthocyanin retentions of 26.81% and 17.32% at 37 °C after 30 and 60 days, respectively, whereas anthocyanins encapsulated in SPI–gum Arabic had 60.26% and 48.32% retentions after the same respective periods. In contrast, Khazaei et al. [55] investigated the storage stability of anthocyanins from saffron petals that were microencapsulated using maltodextrin and gum Arabic conjugates. The outcome revealed no significant difference in the protective effect when using a single wall material and various compositions over 10 weeks at 35 °C.

The DE of maltodextrin is of paramount importance in the stability and physical properties of pigments, and it has been investigated in the freeze-dried microencapsulation of anthocyanins. Maltodextrin with a lower DE might lead to the production of powder with lower water absorption due to the lower number of hydrophilic branches than maltodextrin with a higher DE [56]. Nevertheless, a higher DE might enhance the stability of anthocyanins because of the superior T_g [6]. Moreover, it has been stated that as the concentration of higher-molecular-weight chains increases (lower DE), maltodextrin acts like starch, resulting in high cohesiveness and viscosity. However, a higher DE maltodextrin (with shorter chains) enhances the browning, hygroscopicity, and solubility. Celli et al. [57] investigated the influence of maltodextrin with various DEs (4.0–7.0 (M7), DE 13–17 (M17), and DE 16.5–19.5 (M19)) on the storage stability and physical properties of microencapsulated lowbush blueberries. Anthocyanins encapsulated using M7 led to a brittle and flaky powder with a heterogeneous appearance, both dark and pale pink. Furthermore, they had a wide range of particle distribution and lower bulk density. However, M17 and M19 samples resulted in a homogeneous and glass-like appearance with a narrow size distribution and a higher bulk density. Importantly, the maltodextrin with a higher DE demonstrated better performance in slowing the anthocyanins' degradation. The authors estimated that half of the anthocyanins would be degraded after 8.3 years, 3.8 years, and 10.5 months for M19, M17, and M7, respectively, at 45 °C. Jafari et al. [58], who microencapsulated saffron petal anthocyanins in maltodextrin with different DEs (20 and 7), cress seed gum, and gum Arabic, found that various formulated powders did not experience a drastic change in total anthocyanin content. Moreover, the ability to protect the formulations, irrespective of the use of single or double wall materials, was almost the same during ten weeks of

storage at 35 °C. According to Yamashita et al. [59], maltodextrin with 10 DE resulted in better physical properties regarding color indices, aw, and hygroscopicity. It also gave an anthocyanin retention of around 297.86 mg cyd 3-glu/100 g db, but MD 20 DE led to nearly 265.73 mg cyd 3-glu/100 g db for blackberry byproducts. These contradictions among the findings of different studies make freeze-dried microencapsulation of anthocyanins a complicated method.

Nevertheless, freeze drying is considered the least damaging approach to encapsulating anthocyanins; however, it is time-consuming and requires high cost and energy. Another drawback of this method is the production of the open porous structure powders, making them sensitive to oxidation if the product is not packed under inert atmospheric or vacuum conditions. Furthermore, most freeze-dried powders only provide stability upon storage and not in the gastrointestinal tract since the high porous wall provides inadequate protection for prolonged release [16].

2.1.3. Electrospraying/Electrospinning

Electrohydrodynamic processes are composed of two sister technologies, including electrospraying and electrospinning, and apply high-voltage electrostatic forces to fabricate electrically charged jets from a polymer solution. There are only minor differences between the two methods. Electrospraying is referred to as the formation of solid particles once the solution has a low concentration and viscosity. However, a transition from electrospray to electrospinning occurs by using higher concentrations and viscose solution, which cause entanglements and cohesion of the molecular chain, resulting in nano- or microscale ultrathin continuous fibers. These processes can be divided into uniaxial and coaxial. In the former, core and wall materials are pumped into the electric field via the same tip. Still, in the latter, the wall material solution and bioactive ingredients are simultaneously ejected through two various concentric nozzles. Finally, the inner liquid is encapsulated by an outer polymeric solution [60,61]. These methods provide a broad range of benefits. They are innovative, cost-effective, and one-step methods that offer a promising approach for the fabrication of encapsulated bioactive ingredients. Moreover, they are performed at atmospheric pressure and room temperature, which are suitable conditions for the encapsulation of heat-sensitive components and protecting them from thermal degradation [62]. Importantly, they result in a high surface-to-volume ratio, enhanced EE, tailored release of bioactive molecules, and improved functionality and physical properties of the composites. Additionally, they do not utilize chemical crosslinkers and do not require postprocessing treatments, hence taking priority over the aforementioned methods [17]. The EE, morphology, and physical properties of particles or fibers are mainly influenced by operational parameters, ambient conditions, and intrinsic properties of the solutions. Processing conditions consist of the flow rate, electrical potential applied, and distance between the tip of the syringe nozzle and the collector. Ambient conditions are modified in terms of the humidity and temperature. Intrinsic properties also include the surface tension, viscosity, concentration, molecular weight of polymer, and electrical conductivity.

Dicastillo et al. [63] employed an electrohydrodynamic process for the encapsulation of açaí fruit extract into zein electrosprayed particles. The formation of ultrathin fibers was reported while applying zein (20% *w/v*). However, the electric charge broke the chains while using zein (16% *w/v*), resulting in dispersed droplets. They effectively performed encapsulation with an efficiency of around 72%. Furthermore, the phenolic loss of the encapsulated samples was two and eight times lower than commercial dehydrated açaí fruit after baking (180 °C) and sterilization (121 °C), respectively. Moreover, these authors reported that as the injection flow increased, the solvent could not entirely evaporate, causing aggregation and producing particles that were larger in diameter. In another study, Atay et al. [64] incorporated black carrot anthocyanins into a blend of gelatin and chitosan through the electrospraying technique. They emphasized the total solid content and rheological properties of the solution in capsule formation, according to which, an increased concentration led to a higher viscosity that hindered the proper evaporation

of the solvent. At the same time, a decreased concentration caused insufficient chain entanglements. It was reported that capsule development only occurred when the range of solid content was between 6.4 and 6.8%.

In the study by Isik et al., anthocyanins from sour cherry were encapsulated in gelatin and gelatin–lactalbumin using coaxial and uniaxial electrospraying. According to spectrophotometric results, the EEs varied between 70.2 and 79.2%. Moreover, the bioaccessibility of the coaxially encapsulated sample was eightfold higher than the nonencapsulated one. In another study, Dicastillo et al. [65] produced hydroxypropyl-β-cyclodextrin electrospun microcapsules that were incorporated using different concentrations of maqui extract varying between 10 and 40% (*v/v*). It was found that the viscosity of the solution was affected by the amount of added extract and morphology and the particle size of the spun capsules turned from a spherical to a fiber-like structure by applying concentrations over 30% (*v/v*) of the extract. Furthermore, the phenolic compounds were well preserved in electrospun cyclodextrin capsules after being exposed to baking and autoclaving temperatures.

The optimal electrical conductivity of the solutions is another factor regarding assuring successful electrospinning/electrospraying. Sufficient electrical conductivity is required to create a repulsion force and ultimately lower the droplets′ surface tension to develop a polymer jet. Despite this, superior electrical conductivity poses difficulties since the solution does not possess adequate charge repulsion to overcome its surface tension [66]. Several studies have reported an increase in conductivity upon incorporating anthocyanin into the polymer solution and higher elongation of jet and fibers with a smaller diameter [67–69].

Although there are several benefits of electrohydrodynamic processes, the fact that they are slow, time-consuming, and produce a low encapsulation yield should not be considered trivial. Moreover, the production of encapsulated samples with desired physicochemical and morphological properties is still a challenge, meaning that the method is hardly reproducible, particularly on an industrial scale. Moreover, the number of studies that have used electrohydrodynamic processes for the encapsulation of anthocyanins is extremely limited, highlighting the need for a thorough investigation of the effect of the process parameters on the EE and other physicochemical properties of the particles or fibers.

2.2. Chemical Methods

Inclusion Complexation

The formation of inclusion complexes is considered to be an encapsulation method in which a bioactive guest compound or part of the molecule can fit into a cavity-bearing host molecule via van der Waals forces, hydrogen bonding, or hydrophobic interactions [70]. Cyclodextrins (CDs) are the most widely used materials for forming inclusion complexes with an extensive range of organic substances. CDs are a series of naturally accruing cyclic D-glucopyranose oligomers possessing six (α-CD), seven (β-CD), or eight (γ-CD) glucopyranose units linked by α-(1,4)-glucosidic bonds, leading to the formation of a torus-shaped ring structure. Depending on the type of CDs, they have different cavity diameters, which are approximately 6, 8, and 10 Å for α-CD, β-CD, and γ-CD, respectively. Nevertheless, the depth of the cavities for the three CDs is the same (≈7.8 Å) (Figure 3) [71]. The internal part of the CDs has nonpolar nature, making them favorable for hydrophobic components, whereas the hydrophilic exterior surface provides polarity and accelerates the water-solubility of guest molecules [72]. The formation of inclusion complexes and their stability largely depends on the shape/size fit between the guest molecule and the host CD. Consequently, various sorts of CDs would present different inclusion complexation capacities with the same guest molecule, leading to dramatically different biological and physicochemical properties [73]. Among all the CDs, β-CD is the most prevalent material used in the food sector due to its availability, safety, and fair price. The diameter of the β-CD cavity is nearly 7.8 Å, and the molecular size of natural anthocyanin is about 6.3 × 12 Å; thus, benzopyrylium or phenyl moiety of the anthocyanin can form a complex with β-CD [74]. In other words, the cavity and hollow molecular shape of the CDs make

them ideal candidates to decrease anthocyanins loss by preserving them from polymerization, oxidation, and degradation reactions. This characteristic can also provide thermal stability and controlled release properties [5]. In this direction, Popović et al. [75] showed that β-CD could prolong the release behavior of cornelian cherry up to 50% over the first two hours. Furthermore, the degradation temperature shifted from 219 °C for free cornelian cherry to 225 °C for cornelian β-CD/cornelian cherry. Li et al. [76] examined the light stability of β-CD microcapsules containing mulberry anthocyanins and displayed 1.36 times higher anthocyanin retention for microcapsules than nonencapsulated anthocyanins. Additionally, the microcapsules' storage stability was nearly 9% and 7% higher than using solely anthocyanin within 28 days of storage under unpackaged and vacuum packaged conditions, respectively. Fernandes et al. [77] also demonstrated the efficiency of β-CD at reducing the blackberry pigments' degradation rate under a simulated gastrointestinal environment. Ahmad et al. [78] compared the EE, color, and morphological properties of saffron-anthocyanin-loaded β-CD and β-glucan and observed superior values for the EE and a whiter powder in the case of β-CD. In terms of the size and morphology, β-glucan formed spherical capsules that were smaller in size, while β-CD formed particles with uneven shapes and various orientations.

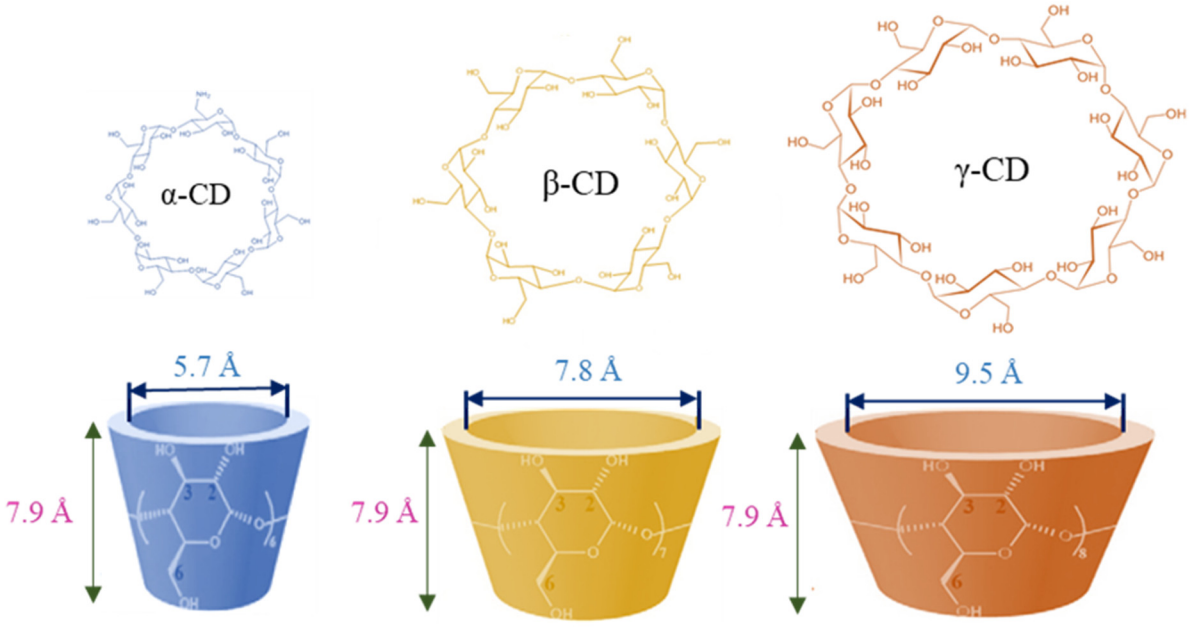

Figure 3. Molecular structure and dimensional characteristics of various cyclodextrins [71].

Anthocyanins in the form of flavylium cations are poorly protected in the cyclodextrin cavity because of their planar structure and polarity. In contrast, more flexible colorless and neutral forms are better candidates for forming inclusion complexes. Howard et al. [79] indicated that incorporating 3% β-CD into natural chokeberry with a pH of 3.6 led to the remarkable retention of anthocyanins, with 95% and 81% protection over eight months of storage at 4 °C and 25 °C, respectively. However, the protective effect was not recorded at pH 2.8, stressing the importance of the anthocyanin structure in β-CD stabilization.

Despite this approach's benefits, β-CD displays low solubility in water, roughly 1.85 g/100 mL, mainly because of its hydrogen bonding, resulting in heightened viscosity at high concentrations or its precipitation. Li et al. [76] reported the improved EE of mulberry anthocyanin–β-CD microcapsules by increasing the core:wall material ratio from 1:2 to 1:6. However, poor dispersion of β-CD hindered the increase in the EE when the proportion exceeded 1:6. To address this issue, Jung et al. [74] introduced cycloamylose (CA) as an emerging complexing agent, which was capable of producing a stable complex at high concentrations due to having a different cavity geometry and outstanding solubility of

>100 g/100 mL. They observed a drastic reduction in the thermal and photodegradation rate constants of anthocyanins while applying CA instead of β-CD, even at high concentrations.

2.3. Physicochemical Methods

2.3.1. Emulsions and Double Emulsions

Emulsions are often used as a medium to enable the wide use of various components that do not dissolve in an aqueous environment. Emulsions are an excellent means of distributing functional substances in aqueous media, protecting them from degradation and greatly influencing their release control.

One of the continuously developed areas in the field of emulsion application is Pickering emulsions. The most frequently claimed advantage of this type of emulsion is that no surfactants are needed, droplets are avoided, and it is very stable. For example, Zhou et al. [80] showed that the combination of gliadin and proanthocyanins as the stabilizer for Pickering emulsions enhanced the antioxidant activity of the emulsion. In a study by Ju et al. [81], an anthocyanin complex with black rice and soy protein isolate was used to produce Pickering emulsions. The results showed that the emulsions stabilized by the complex of anthocyanin nanoparticles and soy protein isolate had very high stability while reducing the oxidation of soybean oil, which was the core material in the emulsion. Interestingly, the slower digestion rate of Pickering emulsions was also demonstrated, making it possible to use this type of product in food with a reduced caloric value.

Another type of emulsion that is often encountered when using anthocyanins is the double emulsion, most often of the water-in-oil-in-water (W/O/W) type. A double emulsion of water-in-oil-in-water (W/O/W) is a microencapsulation procedure, which is used to gain easily controlled release as a carrier for hydrophilic active substances, such as anthocyanins and procyanidin [82]. A double emulsion is, however, not stable, and it is challenging to preserve its composition. Complex coacervation was used in previous works to enhance the double emulsion's efficacy through the electrostatic attraction of oppositely charged biopolymers, allowing for complex coacervated surfaces with reduced solubilization and isolation of the liquid–liquid process [6]. There are various studies involving the application of emulsions for the microencapsulation of anthocyanins. One study used a combination of polyphenols and anthocyanins derived from blueberry that was stabilized using whey proteins to produce double emulsions. The results showed that it is possible to produce optimal emulsions using high-pressure homogenization [83].

Another study focused on the use of guar gum and a double emulsion to enhance anthocyanins' thermal stability. The impacts of various guar gum concentrations on color stability, anthocyanin concentration, and antioxidant capacity were assessed after ten days of storage at 40 °C in the presence of light. According to the researchers, the addition of guar gum had a substantial impact on the samples' antioxidant potential. The double emulsion demonstrated a high encapsulation efficiency and kinetic stability under the conditions tested in this research and shielded anthocyanin molecules from degradation [84]. Another study examined the properties and in vitro release kinetics of anthocyanin-rich microcapsules that were prepared by spraying and freeze drying complex coacervated double emulsions using gelatin–acacia gum (GE–AG) and chitosan–carboxymethylcellulose (CS–CMC). The results indicated that microcapsules might be used to load anthocyanins as a nutraceutical with a managed release requirement [85]. The application of sodium carboxymethylcellulose (CMCNa) to boost the stability and bioavailability of anthocyanin-containing W1/O/W2 emulsions was presented in another study. The emulsions were produced using ultrasound technology, with the inner aqueous phase containing polyglycerol polyricinoleate and the outer aqueous phase containing lecithin and Tween 20. The processes were physiochemically defined over time, and their activity was examined under simulated gastrointestinal conditions. The researchers concluded that the use of CMCNa resulted in high encapsulation efficiencies of more than 90% and improved bioaccessibility [86]. Furthermore, applying the polymer to the lecithin-stabilized emulsions delayed the release of free fatty acids and enhanced the oil digestibility.

The results can be used to improve the design for anthocyanin-emulsion-based delivery systems, ensuring that they operate in food matrices and through the gastrointestinal tract.

2.3.2. Liposomes

When considering aspects such as flavor preservation and release, improved stability, and shelf life of sensitive materials, the microencapsulation technique provides exciting possibilities for food technologists. A new microencapsulation concept involves the liposome microencapsulation of food ingredients that benefit from cheap, commonly accepted as safe substances. Liquid particles, solid particles, or gaseous materials are packed inside continuous shells that can, under desired conditions, release their components at controlled rates. A liposome (or lipid vesicle) is a structure that consists of several aqueous or liquid compartments that enclose lipid bilayers [87]. Over the past 20 years, liposomes have been researched extensively as model membranes and drug delivery systems. Techniques for liposome microencapsulation have been produced, even to the point that they can be used in many commercial implementations. Previously, there have been engaging experiments in anthocyanins' microencapsulation to apply liposomes in the food processing industry. The majority of encapsulating methods are based on the first formation of particles of the active agent, enclosed by a membrane via the various processes [88]. It is possible to think of the liposome as a hollow sphere, whose size can vary from a few nanometers to a few microns. Phospholipid molecules, which are the same molecules that form the cell membrane, are usually made of liposomes. There are various studies involving the application of liposomes for the microencapsulation of anthocyanins.

For preserving anthocyanins, liposomes are a strong and advanced encapsulation vehicle. Liposomes are nano/microsized phospholipid bilayer colloidal vesicles that can be used to encapsulate high-value hydrophilic and hydrophobic components. Liposomes can shield unstable and active compounds from environmental conditions while also increasing their absorption and bioavailability due to their biocompatibility, amphiphilicity, nontoxicity, and non-immunogenicity. An enhanced supercritical carbon dioxide (SC-CO_2) method was used to create anthocyanin-loaded liposomes in a study. Anthocyanin and cholesterol concentrations were investigated. The study's results showed that anthocyanin-loaded liposomes have significant promise in functional food and nutraceutical implementations [87]. Protein–polysaccharide interrelationships open up possibilities for creating new functional foods for food and pharmaceutical applications. One study used a single-stage SC-CO_2 method to develop anthocyanin-loaded liposomes. A phospholipid/anthocyanin suspension equilibrated with CO_2 was depressurized at a constant pressure and rate in this study. This study investigated the effects of strain, depressurization rate, and temperature on the liposome characterization. According to the researchers, this approach allows for the use of dense phase CO_2 to process phospholipid aggregates into nano/microparticles and regulate their properties through the tuning of processing parameters [89]. The SC-CO_2 method had a promising demonstration in the versatile processing of liposomes containing various bioactive compounds for food applications. Anthocyanin encapsulated in liposomes can be shielded from hazardous environmental factors, potentially improving their potency and health advantages in food and nutraceutical compositions.

Liposomes are increasingly attractive for use in food science as a method of encapsulating anthocyanins because they can protect and retain hydrophilic bioactive compounds. Additionally, what also attracts the research attention is the possibility of using liposomes as suitable substance carriers through intestinal absorption with the minor use of carrier agents [90]. However, it is worth paying attention to the fact that liposomes are systems of relatively low stability since the interfacial connections are delicate. Moreover, they do not protect themselves against oxidation because part of the membrane is unsaturated fatty acids. For this reason, the use of antioxidants is even more needed in this matrix.

2.3.3. Ionic Gelation

Ionic gelation is a critical technique among the many methods that are used for encapsulating active compounds because it can be technically regarded as low cost and does not require advanced equipment, high temperatures, or organic solvents. Ionic gelation is a type of microencapsulation that can be achieved using processes of atomization, dripping (coextrusion, extrusion), or electrostatic spraying. This process has the benefit of using mild conditions because high temperatures, intense stirring, or organic solvents are not employed to allow for the encapsulation of substances that, under several conditions, would decay. A downside is that the encapsulation of hydrophilic or low-molecular-weight materials demonstrated issues of easy diffusion and rapid release, irrespective of the pH, via the ionic gel matrix [19]. Some techniques (i.e., emulsion method, coating material) need to be applied to maintain the hydrophilic active compounds because only hydrophobic or low-solubility active compounds are directly applicable to ionic gelation. Several authors have successfully employed the ionic gelation strategy for anthocyanin encapsulation. The larger size and low consistency of particles were noted by the authors as being unfavorable points when using this approach (mainly for hydrophilic active compounds).

The low polydispersity and strong encapsulation quality are the desirable aspects. In various independent studies with different preparation methods and core materials, macro- and microparticles have been examined. Therefore, in a single sample, the examination of macro and microparticles with coherent conditions will more clearly show the particles' characteristics. It is easier to treat encapsulated bioactive compounds and provides increased stability. Ionic gelation is a fascinating encapsulation process that benefits from ease of use and practicality, while avoiding high temperatures and organic solvents. Particulate types of gel have many usages, including structuring, strengthening, and texturizing agents in food matrices and the potential to enhance visual product acceptability. They can also change shape and scale, allowing for the controlled release of active ingredients in agricultural, pharmaceutical, and food products. There are various studies involving the application of ionic gelation for the microencapsulation of anthocyanins.

Moura et al. [91] conducted a recent study on the encapsulation of anthocyanin extract from hibiscus calyces with the double emulsion and two ionic gelation techniques (dripping–extrusion and atomization) and evaluated the stability of the resulting microcapsules at various storage temperatures. The authors concluded that encapsulation using the dripping–extrusion technique achieved the most negligible anthocyanin degradation under refrigerated storage conditions, with the encapsulation efficiencies ranging from 67.9 to 93.9%. Another study used the ionic gelation process with dripping–extrusion and atomization to create microparticles comprising anthocyanin extract from Hibiscus sabdariffa L. (HE). A crosslinked solution ($CaCl_2$) and a double emulsion (HE/rapeseed oil/pectin) were used. The goal of the research was to see how anthocyanins were released under simulated gastrointestinal environments and how microparticles were used in pectin candy [19]. The authors concluded that the microencapsulation of hibiscus anthocyanin enhanced the bioactive compound's enteric defense, primarily in microparticles formed using dripping–extrusion. Anthocyanin encapsulation by the extract was characterized in another study using an adsorption technique with blank alginate beads. Ionic gelation was combined with a complexation process involving chitosan, whey protein concentrate, or gelatin in this study. The use of cationic polymers in the complexation process was effective in preserving these pigments during refrigerated storage. The researchers concluded that the ionic gelation encapsulation process resulted in the creation of hydrogel beads containing anthocyanins, allowing for the analysis of the compounds' release profiles under particular pH conditions, such as those contained in intestinal fluid [92].

2.3.4. Coacervation

Most recently, due to the high loading capacity, low temperature operating requirements, reduced evaporation losses of volatile compounds and thermal degradation, and

greater competence in regulating the release rate of nutraceuticals, there is considerable interest in the coacervation technique among all the microencapsulation technologies.

Coacervates are a spherical aggregate of organic macromolecules that form an inclusion that is held together by hydrophobic factors. Simple organic molecules may be selectively adsorbed from the surrounding medium due to the boundaries of the coacervates. The drop that forms the coacervate particle is frequently a liquid oil. The drops will have a shell of the hydrocolloid mixture surrounding them, which will set to form a solid or semipermeable shell under the right conditions. Since both entropy and enthalpy regulate the coacervate formation, a minimum temperature must be preserved [93]. Coacervation involves forming microscopic droplets of the coacervate phase in a stirred liquid phase, which typically results in an unstable colloidal dispersion with a tendency to coalescence. Crosslinking or mechanical gelation of the polymer in the coacervate process will retain and sustain droplets. Coacervation mechanisms are divided into two categories: basic coacervation and complex coacervation [94]. Simple coacervation is commonly used in medical technology to entrap drugs into microcapsules. There are various studies involving the application of coacervates for the microencapsulation of anthocyanins.

In one of the studies, raspberry water extracts were microencapsulated using a double emulsion methodology prior to complex coacervation with gelatin and gum Arabic to reduce their anthocyanins' instability as water-soluble substances, especially under harsh processing and storage conditions. The findings demonstrated microcapsule thermostability [95]. The optimal condition microcapsules retained an intense red color over time, suggesting that the method used to conserve anthocyanins was successful. Regulated protein–polysaccharide interactions through complex coacervation may point to a way to improve their functional position as ingredients without requiring chemical or enzymatic changes.

Another study involving coacervation was a study by Guo et al. [96], who used blueberry- and purple-corn-derived anthocyanins to encapsulate particles based on hydrogels with alginate and pectin. A very high microencapsulation efficiency was demonstrated. At the same time, anthocyanins could be stored longer, thanks to this method.

Like any method, coacervation also has some limitations, mainly related to the agglomeration of particles and particle size control. The second area of disadvantage is that, by their principle, the particles created by this method are very sensitive to pH and ionic strength, limiting their use in various matrices [70].

3. Conclusions

Anthocyanins play a very important role both in human nutrition and in the food industry as colorants. A very important function is played by their ability to change color depending on the pH in which they are located. However, this lability also necessitates their protection against environmental factors. For this reason, increasingly more attention is paid to the role of microencapsulating anthocyanins. The most common method of microencapsulating anthocyanins is still spray drying, followed by freeze drying, because thanks to these methods, high yields are obtained, even at an industrial scale. On the other hand, physical methods, such as electrospinning, and chemical ones, such as inclusion complexation, are gaining increasingly more recognition among scientists. Very interesting applications for the microencapsulation of anthocyanins are obtained using physicochemical methods, such as when using liposomes or ionic gelation. The current review has exhausted the topic of applications of anthocyanin microencapsulation using the most common and modern methods. Possible future recommendations are to apply microencapsulated anthocyanins as food fortification ingredients in real food products and investigate their bioaccessibility, digestibility, and stability. It is also crucial to focus on biopolymers with pro-health value as wall materials to provide extra health benefits. Furthermore, further studies are suggested to investigate the feasibility of combinations of techniques for protecting anthocyanins.

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