

Natural colorants from plant pigments and their encapsulation: An emerging window for the food industry



Susmita Ghosh ^a, Tanmay Sarkar ^{a,b}, Arpita Das ^c, Runu Chakraborty ^{a,*}

^a Department of Food Technology and Biochemical Engineering, Jadavpur University, Kolkata, 700032, India

^b Malda Polytechnic, West Bengal State Council of Technical Education, Govt. of West Bengal, Malda, 732102, India

^c Department of Biotechnology, School of Life Science and Biotechnology, Adamas University, Barasat, West Bengal, 700126, India

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ABSTRACT

Color is considered to be the primary feature perceived by the senses that represent a crucial role for centuries in the acceptability of foods to enhance their actual appearance and quality. Plant pigments, the impeccable natural source of color, display enormous potential to substitute many of the synthetic colorants. Chlorophylls, carotenoids, anthocyanins, and betalains are the extensive classes of natural colors contributing comprehensive color shades to foods. There are good perspectives for the inclusion of plant pigments in the food industry. Their incorporation into food products is very challenging, as they are chemically unstable and exhibit poor bioavailability. Encapsulation is an excellent process to enhance its bioaccessibility, digestibility, and controlled release. During food fortification, efficient encapsulation technologies are needed to prevent the degradation of pigments and reserve their bioavailability in the human gastrointestinal system. The development of cost-effective and viable technologies for the preparation of natural food color and its application in foods is a great challenge and a major need of the day.

1. Introduction

Plant pigments are the unique chemical substances that are responsible for colorful appearances and the visual attraction of fruits and vegetables. They are mainly considered as secondary plant compounds that play essential roles in critical biological processes of plants including metabolism, light-harvesting in photosynthesis, regulation in development and defense, and protection from photo-oxidative damage. Their consumption has been associated with decreased chances of developing various diseases in humans (Ghosh, Sarkar, Das, & Chakraborty, 2021). Color is the most outstanding parameter by which the quality of foods is judged that can stimulate or suppress one's appetite. Food colorants make foods more attractive, appetizing, and recognizable. Artificial colorants are used in foods for a brighter appearance and economic motivation because foods with attractive colors enhance the appeal and stimulate the appetite. Natural colorants have become more popular around the world as a result of their therapeutic and medical effects as well as the high toxicity of synthetic colors (Jurić et al., 2020; Lu et al., 2021). Natural pigments have various problems, such as lack of stability and low bioavailability, despite their coloring qualities and health-promoting actions. Encapsulation technology has sparked an

increasing interest in various disciplines and applications over the last few decades. It provides a comprehensive perspective to the development of novel and healthier foods. It was first developed as a technology to provide a coating to solids, liquids, and gaseous compounds that cause their controlled and targeted release at a specific rate. Bioactive ingredients like pigments or vitamins are protected through encapsulation techniques that provide a shield from deleterious environmental conditions and allows for efficient processing. Natural pigments can be encapsulated in functional foods to extend their health-promoting activities to a wider population (Ghosh et al., 2021). The aim of this review article is to provide a concise illumination of several aspects of natural colorants that include-

- i. A precise overview of the health benefits, stability, bioavailability, and bioaccessibility and application of natural pigments from plant sources.
- ii. The application of encapsulated pigments in the food industry.

This review is focusing on the major concerns surrounding the application of natural pigments from plant sources in foods, beginning with the sources, classification, health-promoting activities, extraction

* Corresponding author.

E-mail address: crunu@hotmail.com (R. Chakraborty).

techniques, need for encapsulation, different processes of encapsulation, and the use of encapsulated pigments in a variety of food systems.

2. Natural colorants from plant pigments

Pigments cause the natural spectacular color of plants. Red-yellow betalains, green chlorophylls, red-purple anthocyanins, and yellow-orange carotenoids are the most common pigments found in vegetables and fruits. Apart from their coloring properties, these plant pigments exhibit potential health-promoting functions (Ghosh et al., 2021).

Agricultural residues are a key source of natural pigments that are increasingly being recognised as fundamentally sustainable, and their utilization for natural pigments recovery could help to reduce the environmental difficulties connected with their management. Seeds and peels of different fruits and vegetables, which are removed while processing, may still retain significant levels of pigments, frequently in greater concentrations (Sharma, Usmani, Gupta, & Bhat, 2021).

3. Classification of plant pigments

3.1. Pyrrole derivatives: Chlorophyll

Chlorophylls are the famous photosynthetic greenish pigments, found in algae, plants, and cyanobacteria that belong to a major class of tetrapyrroles. It is one of the most widely distributed natural pigments which act as a key component of photosynthesis. Chlorophyll breakdown is a vital catabolic process of leaf senescence and fruit ripening. It is potentially applicable in various fields of science and industrial technology due to its unique characteristics. Chlorophyll can be categorized into five major classes which are a, b, c, d, and f. Chlorophyll is distributed largely in the plant kingdom including green fruits and vegetables (Jurić et al., 2020). Basic structure of chlorophylls is the porphyrin macrocycle that consists of four pyrrole rings. Magnesium (Mg) is the central metal in chlorophylls (Pareek et al., 2017). In the structure of chlorophyll, one edge of the porphyrin ring is hydrophilic whereas the side chain containing the phytol chain is strongly hydrophobic. This structure is advantageous to the arrangements of chlorophyll in the phospholipid bilayer membrane. Various conjugated double bonds impart an advantage to the ability of chlorophyll to absorb visible light (Singh, Rana, & Pandey, 2020).

3.2. Isoprenoid derivatives: Carotenoids

Carotenoids are widely distributed ubiquitous lipid-soluble natural pigments that produce yellow, red, and orange pigmentation. The natural source of carotenoids includes plants, microalgae, bacteria, and fungi. Based on functional groups, carotenoids can be subdivided into two classes – carotenes which contain only the parent hydrocarbon chain, including α -carotene, xanthophylls, lycopene, and β -carotene containing oxygen as functional groups, such as lutein and zeaxanthin. Oxidation destroys carotenoids whereas geometrical isomerization enhances the proportion of Z isomers. Carotenoids belong to the class of isoprenoid lipids. The formation of color is dependent upon their conjugated carbon–carbon double bonds in the chemical structure. The system of alternating double and single bonds constitutes a conjugated system (Shilpa, Shwetha, Raju, & Lakshminarayana, 2020).

3.3. Flavonoids derivative: Anthocyanin

Flavonoids belong to the class of polyphenolic compounds that have a 2-phenylchromane nucleus and demonstrate excellent therapeutic, pharmacological, and biochemical functions. Anthocyanins are the water-soluble pigments in the flavonoids group that exhibit red, purple, and blue colors plants, and display crucial roles in plant propagation and plant defense mechanisms. Anthocyanins are glycosylated, polyhydroxy, or polymethoxy derivatives of flavylium cation ($C_{15}H_{11}O^+$). The

molecular weight of flavylium cation is 207.24724 g/mol and it has a positive charge at the oxygen atom of the C-ring of the basic flavonoid structure. Most commonly distributed anthocyanidins in the plants include cyanidin, malvidin, petunidin, delphinidin, pelargonidin, and peonidin. The formation of red, blue, and purple coloring plants is dependent upon the conjugated bonds of anthocyanins (Eker et al., 2020).

3.4. Nitrogen-Heterocyclic derivatives: Betalains

Betalains are tyrosine-derived water-soluble nitrogenous plant pigments and they consist of a betalamic acid [4-(2-oxoethylidene)-1,2,3,4-tetrahydropyridine-2,6-dicarboxylic acid]. Betalains can be classified into two major classes: the yellow-orange betaxanthin and the red-violet betacyanin. Betalains are present in edible portions of the plants and the leaves, flowers, and stems. Betanin is the omnipresent betacyanin among plants and red beet is the most common source of betanin. Vulgaxanthin and indicaxanthin are present in the yellow beet and cactus pear. Depending on the pigment proportion, the combined existence of betaxanthins and betacyanins in similar parts of the plants produces orange to red color. Betacyanins and betaxanthins display absorbance maxima centered at wavelengths of $\lambda_m = 536$ nm and $\lambda_m = 480$ nm respectively. In betacyanins, glycosylation and acylglycosylation of the hydroxyl groups are reported whereas no glycosylation has been observed in betaxanthins (Hussain, Sadiq, & Zia-Ul-Haq, 2018).

4. Extraction of Pigments

To increase the efficiency and production of natural colorants, an appropriate extraction procedure for the natural pigments must be selected. Extraction of crude pigments and other chemicals is frequently done using traditional methods. Non-traditional extraction methods, often known as green extraction techniques, have recently emerged as viable alternatives to conventional extraction since they use less solvent and take less time. Soxhlet extraction, maceration, and hydrodistillation are examples of traditional processes that are simple, economical, and straightforward to use (Ngamwonglumlert, Devahastin, & Chiewchan, 2017a). Other emerging technologies, such as ultra-high pressure, negative pressure cavitation, high voltage electrical discharges, ohmic heating, pulsed electric fields, mechano-chemical methods, and high-pressure homogenization, have proven to be very efficient methods for the extraction of plant pigments. Furthermore, the consumption of extracted pigments has been associated with human health promoting effects (Jurić et al., 2020; Ngamwonglumlert, Devahastin, & Chiewchan, 2017b).

5. Need for encapsulation of pigments

5.1. Stability of pigments

Chlorophyll is a very unstable compound and the stability of chlorophyll is highly affected by pH, temperature, heat, and light. Chlorophyll is more thermolabile than carotenoids. Microwave and conventional heating of kiwi result in a loss of 42–100% in the chlorophylls (Benloch-Tinoco et al., 2015). During the processing and storage of foods that result in color loss, chlorophylls are highly susceptible to degradation. The chlorophyll content decreased faster under light than in the dark because of singlet oxygen. Samples with added lipids showed lower and slower degradation of chlorophyll than in samples without lipids. High pressure high temperature processing results in the degradation of chlorophyll *a* and chlorophyll *b*. Both chlorophylls were highly degraded at 117 °C (Sánchez, Baranda, & De Marañón, 2014).

During the storage and processing, carotenoids in foods are destroyed because of oxidation. Carotenoid pigments are sensitive to pH, temperature, heat, and light. When carotenoids are thermally treated, it provides volatile compounds and larger non-volatile

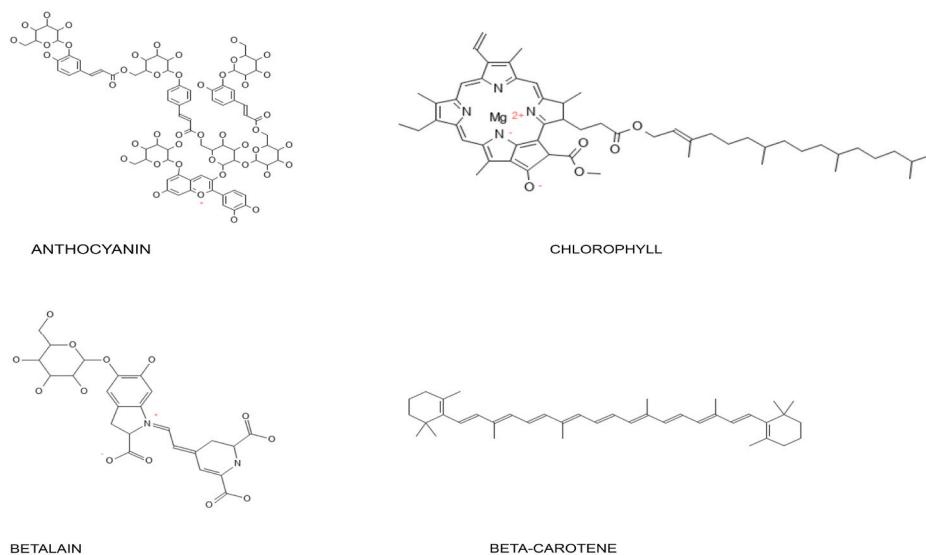


Fig. 1. Structure of different natural pigments.

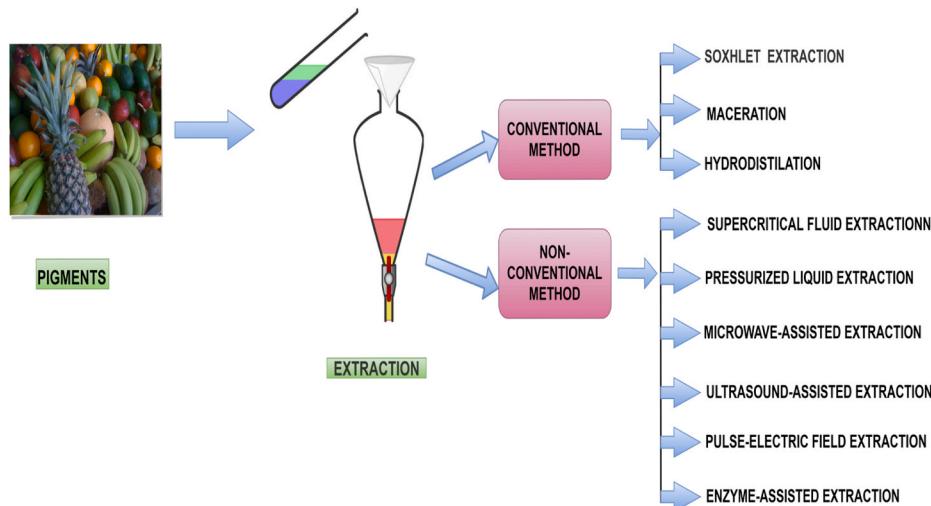


Fig. 2. Extraction of pigments.

compounds. Light exposure results in the degradation of carotenoids. The primary causes of carotenoids degradation are oxidation and isomerization that results in loss of color. Heat, light, and acids influence the isomerization of carotenoids from *trans*-isomer to *cis*-isomers (Lu, Maidannyk, & Lim, 2020). The application of anthocyanins in the food and pharmaceutical industries is limited as they are very much unstable and prone to degradation. Temperature, pH, oxygen, light, metal ions, and enzymes greatly influence the stability of anthocyanins. A high pH value destroys anthocyanins. In comparison with other co-pigments, caffeic acid exhibits more stability than anthocyanins. Low pH and low temperature exert a positive impact on the higher level of anthocyanins stability (Liu et al., 2018).

The stability of betalain is chiefly affected by temperature, the concentration of pigment, pH, co-presence of compounds, water activity, oxygen, light, chelating agents, storage and processing conditions, and enzymes. Betalains are heat-sensitive molecules and temperature level and heating period affect the rate of degradation. At the optimum pH level, thermal degradation of pH results in color loss or browning. Water activity is a crucial factor for the stability of betalain because of water-dependent hydrolytic reaction. Exposure to daylight at 15 °C results in degradation of betalain pigment up to 15.6% (Hussain et al., 2018).

5.2. Bioavailability and bioaccessibility of pigments

Natural pigments have a high level of antioxidant activity and provide a variety of health benefits. Bioaccessibility and bioavailability determine whether these beneficial bioactive chemicals can be successfully employed by the human body. Bioaccessibility refers to the percentage of ingested natural pigments that are absorbed by the intestine, while bioavailability refers to the total amount of natural pigments that are absorbed by the gut. Bioaccessibility and bioavailability of natural pigments are determined by their principal digestive processes. Their bioaccessibility and bioavailability are also influenced by their chemical characteristics (Lu et al., 2021).

Chlorophyll structure and food matrix mainly limit chlorophyll bioaccessibility or bioavailability. In different seaweed species, chlorophyll exhibits various micellization and absorption rate. For determining chlorophyll bioaccessibility, the characteristics of the food matrix are one of the most important parameters (Lu et al., 2021). The absorption of carotenoids is dependent on their bioaccessibility to a great extent. Lipids enhance the bioaccessibility and absorption of carotenoids. Carotenoids obtained from plant foods get absorbed inefficiently. Food processing, digestive enzymes, the composition of the

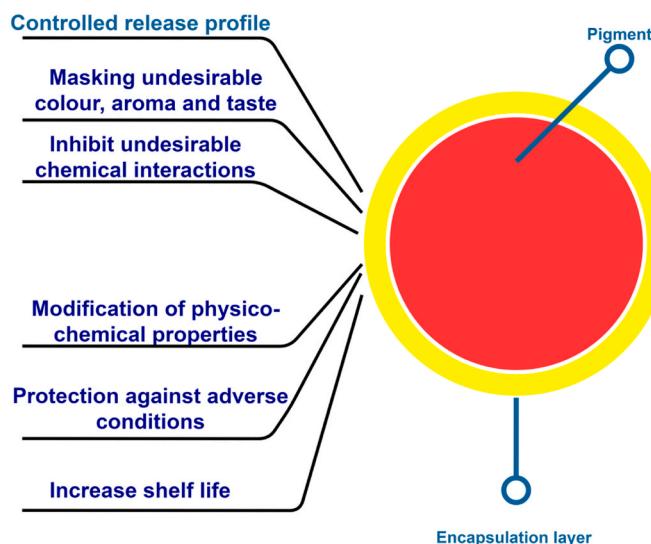


Fig. 3. Need for pigment encapsulation.

meal, the efficiency of transport across the enterocytes affect the absorption of carotenoids. Various dietary factors that influence the bioavailability of carotenoids are the type and quantity of lipids, fibers, micronutrients, and phytochemicals. Some non-dietary factors that have an impact on the bioavailability of carotenoids include age, gender, physiological status and disease, and absorption of fat (Shilpa et al., 2020).

The bioaccessibility of anthocyanin indicates availability for absorption. Cell metabolism and translocation across the basolateral membrane chiefly influence the bioavailability of anthocyanin. Anthocyanins with fewer hydrophilic groups exhibit low bioavailability and anthocyanins with more hydrophilic groups represent excellent bioavailability. Food processing and food matrix, digestive enzymes, pH, gut microbiota, motility, and permeability of the GI tract - are the predominant considerations that influence the alterations in the bioavailability of anthocyanin. Metabolites obtained from microbial metabolism are the main anthocyanin forms in the circulation and exert beneficial health effects (Eker et al., 2020).

The food matrix limits the bioaccessibility of betanin. Food matrix

could prohibit the degradation of betanin and isobetanin. The bioaccessibility of betaxanthins is greater than betacyanins. The bioavailability of Indicaxanthin is greater than betanin. The stability of dietary betalains is linked with its bioavailability. Betalains are stable at a moderate level at the in vitro state of the gastrointestinal tract. The low bioavailability of betanin is related to its post-absorptive dissemination in the different compartments of the body (Hussain et al., 2018).

6. Encapsulation of pigments

Natural pigments are a safe food additive to add natural color and display a promising potential to promote health and prevent diseases as therapeutic agents. The best option to stabilize and enhance the application of plant pigments derives from their encapsulation. Encapsulation is a burgeoning field of research that opens up new scopes for the development of more developed and technologically advanced foods in the twenty-first century. Improved thermal and chemical stability, preservation or masking of flavor, taste, or scent, controlled and targeted release, and increased bioavailability of pigments are all advantages of encapsulation. Pigment encapsulation at the micro and nanoscales will give extensive and intense platforms for the development of a new stage in the production of novel and healthful foods. Microencapsulation and nanoencapsulation are effective platforms for protecting pigments from harmful environmental conditions while also allowing for controlled and targeted release. Nanoencapsulation has recently played an important role to determine the way for the advancement of bioactive and therapeutic supplement delivery in the food industries. There are different techniques for the nanoencapsulation of food ingredients including emulsification, inclusion, coacervation, and complexation nanoprecipitation (Ghosh et al., 2021).

7. Different Encapsulation Techniques involved in the encapsulation of pigments

Microencapsulation by spray drying is a process that helps to maintain pigments after including a coating or carrier material that preserves and facilitates their absorption. It is effective in producing microencapsulated compounds from natural sources containing a variety of colors, including carotenoids, anthocyanins, betalains, and chlorophylls (Carmona, Robert, Vergara, & Sáenz, 2021). The use of supercritical fluids as an antisolvent has been used to analyze recent

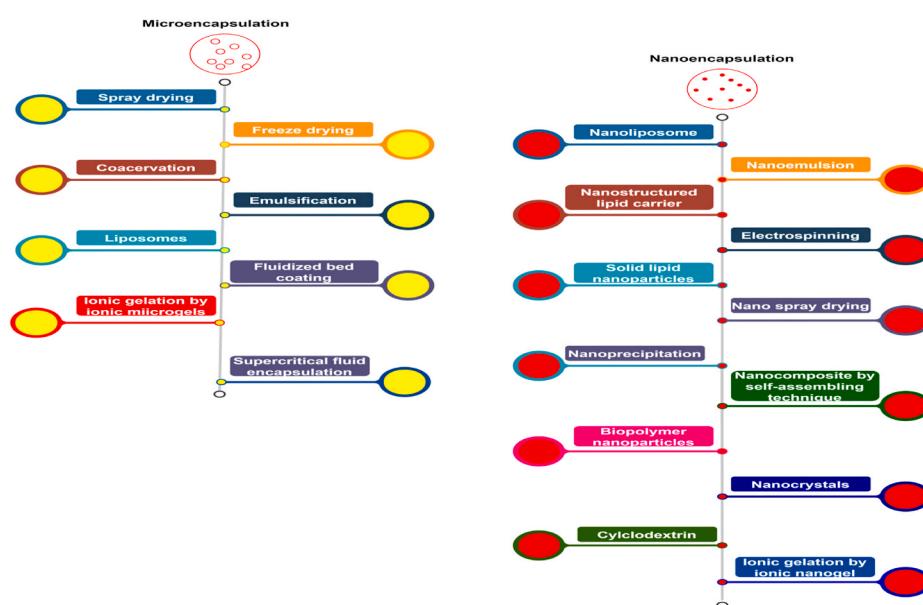


Fig. 4. Different techniques of pigment encapsulation.

Table 1

Major plant pigments –color, subgroup, solubility, chemical structure, stability, bioavailability, and bioaccessibility of pigments.

Plant Pigment	Color & Subgroup	Solubility & Chemical Structure	Application in the food industry	Stability	Major Sources	Factors Affecting Bioavailability & Bioaccessibility	Reference
Chlorophyll	<ul style="list-style-type: none"> green 6 types - a, b, c, d, e, f <p>The main types are chlorophyll <i>a</i> and chlorophyll <i>b</i></p>	<ul style="list-style-type: none"> Soluble in lipid porphyrins in which the pyrrole rings are connected by methyne bridges and the double bond system with a centrally located magnesium atom, coordinated with the nitrogen of the four pyrroles 	Beverages, fruit juices, pasta, dairy products, soups, sweater preparations	<p>Chlorophyll degradation is caused by -excess light, prolong heating, presence of oxygen, under acidic environment, and by enzymes, light, and storage at low-temperatures</p> <ul style="list-style-type: none"> Acylation of sugar moieties, glycosylation of the anthocyanin enhance stability. Ascorbic acid, enzyme (β-G anthocyanase) causes anthocyanin degradation. They are susceptible to temperature, light, solvent, pH, oxygen, presence of proteins. 	Photosynthetic organisms including plants (spinach, broccoli, pepper, green beans, peas), blue-green algae	<p>Chlorophyll structure and food matrix limit chlorophyll bioaccessibility or Bioavailability.</p>	(Jurić et al., 2020a; Singh et al., 2020)
Anthocyanin	<ul style="list-style-type: none"> Dark blue to red Glycosides or acylglycosides of six aglycone anthocyanidins: pelargonidin, cyanidin, delphinidin, peonidin, petunidin, malvidin 	<ul style="list-style-type: none"> Water-soluble flavylium (2-phenylbenzopyrylium) structures with varying hydroxyl and methoxyl substituents 	Bread, cakes, cookies, Jelly and yogurt red pigments, beverages, kefir and carbonated water, Fruit juice	<ul style="list-style-type: none"> •Acylation of sugar moieties, glycosylation of the anthocyanin enhance stability. •Ascorbic acid, enzyme (β-G anthocyanase) causes anthocyanin degradation. They are susceptible to temperature, light, solvent, pH, oxygen, presence of proteins. 	Purple grapes, cherry, plum, raspberry, strawberry, blackberry, blueberry, cranberry, chokeberry, red cabbage	<ul style="list-style-type: none"> Anthocyanins with more hydrophilic groups present excellent bioavailability. Major factors affecting alterations in the bioavailability include Food processing and food matrix, digestive enzymes, pH, gut, and permeability of the GI tract. 	(Eker et al., 2020; Jurić et al., 2020b)
Betalains	<ul style="list-style-type: none"> Yellow to violet Betacyanins and betaxanthins 	<ul style="list-style-type: none"> Water-soluble Immonium conjugates of betalamic acid with cyclo-dopa [cyclo-3-(3,4-dihydroxyphenylalanine)] and amino compounds 	Burgers, desserts, ice cream, jams, jellies, soups, sauces, sweets, drinks, dairy products, yogurts, gummy candy, cow milk	<ul style="list-style-type: none"> Betalain stability is enhanced by Increased pigment content, decreased pH, chelating agents, a greater degree of glucosylation, and acylation, reduced Aw, antioxidants, darkness, and low temperature. Betalain stability is decreased by degrading enzymes (peroxidase, polyphenol oxidase, glucosidase), a lower degree of glucosylation and acylation, high aw, metal cations, pH < 3 or > 7, high temperature, light, and oxygen. 	<i>Basella rubra</i> or Malabar spinach, cactus pear (<i>Opuntia ficus-indica</i>) and <i>O. Stricta</i> , red-purple pitaya (<i>Hylocereus polyrhizus</i>), and Amaranthus species	<ul style="list-style-type: none"> The food matrix limits the bioaccessibility of betanin. The bioaccessibility of betaxanthins is greater than betacyanins. 	(Hussain et al., 2018, pp. 1–187; Jurić et al., 2020b)
Carotenoids	<ul style="list-style-type: none"> Yellow, orange and red β-carotene, α-carotene, β-cryptoxanthin, lycopene, lutein, and zeaxanthin 	<ul style="list-style-type: none"> Soluble in lipid C40 tetraterpenes/tetraterpenoids formed from eight C5 isoprenoid units 	Cheese, butter, margarine, oils and fats, non-alcoholic beverages, fruit juices, baked products, ice creams, yogurts, sweets, jams, creams, pastries, and jellies	<p>Carotenoid degradation is increased with the deterioration of the cellular structure of food, increased surface porosity, duration and level of processing conditions, inadequate storage conditions, permeability to O₂, and light.</p>	Green vegetables, tomato, orange fruits and vegetables, corn, red peppers, seaweed	<p>Factors that influence the bioavailability of anthocyanin include -dietary factors including type and quantity of lipids, fibers, micronutrients, and phytochemicals whereas non-dietary factors include host-related factors like age, gender, physiological status and disease, genetic factors, and absorption of fat</p>	(Lu et al., 2020; Shilpa et al., 2020)

Table 2
Application of encapsulated colour in different food model system.

Plant Pigment	Pigment Origin	Method of Encapsulation	Coating Material	Purpose	Food Model System	Result	Reference
Chlorophyll	Alfalfa leaves (<i>Medicago sativa</i>)	Emulsion	Gelatin, agar, oil phase, and deionized water	To evaluate the impacts of the microencapsulation Technique of green pigment of the <i>M. sativa</i> on heated food	Gummy Candy	There are greater levels of resistance of the microencapsulated dye samples against the heating	Raei, Ali, Ardakani, and Daneshi (2017)
Anthocyanin	Barberry fruits (<i>Berberis vulgaris</i>)	Spray Drying	Gum Arabic, Maltodextrin, and Gelatin	<ul style="list-style-type: none"> • To assess the impact of coating agents, temperature, light, and humidity on the stability of spray-dried barberry anthocyanins. • To evaluate the use of encapsulated anthocyanin as a natural food colorant in jelly formation. 	Jelly	Jelly with added 7% encapsulated color exhibited greater results in comparison with commercial jelly containing artificial color	(Akhavan Mahdavi et al., 2016)
Anthocyanin	Grape (<i>Vitis vinifera</i> L.) skin	Spray Drying	Maltodextrin	To evaluate anthocyanin degradation of and the reducing power of apple puree containing encapsulated grape skin phenolics	Apple Puree	Anthocyanin retention decreased to 72% with improved heating intensity.	Lavelli, Sri Harsha, and Spigno (2016)
Anthocyanin	Cabernet Sauvignon grapes (<i>Vitis vinifera</i> L.)	Spray Drying	Maltodextrin, maltodextrin/cyclodextrin, and maltodextrin/arabic gum	<ul style="list-style-type: none"> • To organize the optimal conditions for the extraction of anthocyanin • To assess the stability of anthocyanins encapsulated with various coating materials in the food system. 	Soft Drink	Anthocyanins applied to an isotonic soft drink system displayed first-order reaction kinetics of degradation	Burin et al. (2011)
Anthocyanin	Black bean (<i>Phaseolus vulgaris</i> L.) coat	Molecular inclusion	β-cyclodextrin	To compare the stability of black bean anthocyanin co-pigmented with β-cyclodextrin	Sport beverage	β-Cyclodextrin improved anthocyanins stability under light and storage conditions.	Aguilera et al. (2016)
Anthocyanin	Sour cherry (<i>Prunus cerasus</i> L.) pomace extract	Freeze Drying	Whey and soy proteins	To formulate functional cookies, fortified with encapsulated sour cherry pomace polyphenols	Cookies	There was a slight increase in total polyphenols of while total anthocyanins and decrease antioxidant activity.	Tumbas Šaponjac et al. (2016)
Betanin	Red beet (<i>Beta vulgaris</i> L.)	Nanoliposomes	Lecithin	<ul style="list-style-type: none"> • To evaluate the encapsulation of betanin-loaded nanoliposomes applied in gummy candies. • To assess the physicochemical and sensorial properties of gummy candies. 	Gummy Candy	Liposomal gummy candies showed greater stability and radical-scavenging activity of betanin in comparison with gummy candies containing free betanin.	Amjadi et al. (2018)
Betacyanin	Prickly pear fruits (<i>Opuntia stricta</i>)	Spray Drying	Glucose syrup	<ul style="list-style-type: none"> • To display a standardized process to obtain a powder colorant by spray drying. • To assess the application of the powdered colorants in yogurt and soft-drink, and to study the color stability. 	Yogurt and Soft Drink	High color strength, non-sticky powder colorant was produced with a drying yield of 58%.	Obón et al. (2009)
Betalains	Barbary fig (<i>Opuntia ficus-indica</i> L.)	Spray drying	Soluble fiber [(1-3)(1-4)-β-D-glucan]	To examine the consequences of encapsulated powder on the physicochemical characteristics of extruded cereals.	Extruded cereal	The encapsulated powder had a positive effect on expansion and water absorption properties and color parameters.	Ruiz-Gutiérrez, Amaya-Guerra, Quintero-Ramos, Pérez-Carrillo, and Meléndez-Pizarro (2017)

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Table 2 (continued)

Plant Pigment	Pigment Origin	Method of Encapsulation	Coating Material	Purpose	Food Model System	Result	Reference
Betalains	Beetroot (<i>Beta vulgaris</i> L.)	Freeze-drying	Maltodextrin, gum Arabic, gum Arabic-modified starch, modified starch chitosan, modified starch maltodextrin-chitosan	<ul style="list-style-type: none"> • To investigate the color stability of the encapsulated colorant • To examine its incorporation in a chewing gum model system. 	Chewing gum	Maltodextrin along with gum arabic acted as effective agents for beetroot and saffron coloring extracts microencapsulation by freeze drying.	Chranioti, Nikoloudaki, and Tzia (2015)
Betalains	Basellaceae fruit (<i>Basella rubra</i> L.)	Nanoliposomes	Lecithin	To optimize a method for the preparation of <i>B. rubra</i> fruit juice betalains nanoliposomes	Gummy Candies	During storage zeta potential, particle size, and polydispersity index of Betalain nanoliposomes didn't change significantly	(Sravan Kumar, Singh Chauhan, & Giridhar, 2020)
Betalains	Cactus pear (<i>Opuntia ficus-indica</i>) pulp	Spray Drying	Maltodextrin, cactus mucilage and mucilage-maltodextrin	<p>To obtain the optimal inlet air temperature and pulp: encapsulating agent ratio for yellow-orange cactus pear pulp and to assess the effect of encapsulating agents, on the microparticles stability and microparticles.</p> <ul style="list-style-type: none"> • To encapsulate betaxanthin by spray drying • To assess the stability of pigments in yogurt and a soft drink colored with betaxanthin-rich extract 	Yogurt	Pulp-Maltodextrin system showed higher indicaxanthin stability and less color change stored at 60 °C. H	Carmona et al. (2021)
Betaxanthin	Cactus Pear Fruits (<i>Opuntia ficus-indica</i>)	Spray Drying	Maltodextrin	<ul style="list-style-type: none"> • To encapsulate betaxanthin by spray drying • To assess the stability of pigments in yogurt and a soft drink colored with betaxanthin-rich extract 	Yogurt and Soft Drink	In both yogurt and soft drink, there was great preservation in the dark after 28 days at 4 °C.	Fernández-lópez et al. (2018)
Carotenoids	Rottb fruit (<i>Renealmia alpinia</i>)	Spray Drying	Maltodextrin, and Gum Arabic, and their mixture	To examine the encapsulated pigments and antioxidant compounds and to assess their potential to be applied as a functional coloring agent	Yogurt	The retention of carotenoids was significantly higher with maltodextrina	(Jimenez-Gonzalez, O., Luna-Guevara, J. J., Ramirez-Rodrigues, M.M. et al. 2021)
β-Carotene	β-carotene were obtained from Sigma-Aldrich (St. Louis, MO, USA)	Solid lipid microparticles	soy protein isolate	<ul style="list-style-type: none"> • To microencapsulate lycopene by spray drying by using a modified starch • To evaluate the application of the capsules in the cake. 	Vanilla Ice cream	Ice cream prepared with partial replacement of artificial colorants by BCSLM containing alpha-tocopherol exhibited a more intense color compared with the food system with non-encapsulated β-carotene.	Lima et al. (2016)
β-Carotene	β-Carotene from Sigma (St. Louis, MO, USA)	Emulsion	Hydrolyzed Soya Protein Isolate	To assess the effectiveness of incorporating β-carotene-loaded solid lipid microparticles stabilized with a hydrolyzed soy protein isolate in yogurt.	Yogurt	Physicochemical or the rheological characteristics of the yogurt were not changed with the presence of the lipid microparticles	Molina et al. (2019)
β-Carotene	β-carotene from Sigma (St. Louis, USA)	Liposome	Xanthan and guar gums	<ul style="list-style-type: none"> • To produce β-carotene-loaded liposomes using proliposomes. • To characterize the β-carotene-loaded liposome dispersions stabilized with the gum mixtures to evaluate their incorporation in yogurts. 	Yogurt	β-carotene-loaded liposomes are remarkably able to protect the encapsulated carotenoids during storage.	Toniazza et al. (2014)

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Table 2 (continued)

Plant Pigment	Pigment Origin	Method of Encapsulation	Coating Material	Purpose	Food Model System	Result	Reference
β-Carotene	Trans β-carotene in powder form with 97% purity	Complex coacervation method	Palm oil with chitosan/carboxymethylcellulose	<ul style="list-style-type: none"> • To encapsulate palm oil and β-carotene with chitosan/carboxymethyl cellulose as coating agent by the process of complex coacervation and with chitosan/sodium tripolyphosphate by the ionic gelation technique. • To evaluate the properties of the microparticles in food models. 	Bread and Yogurt	Particles encapsulated with chitosan/carboxymethylcellulose presented excellent release behavior in water and gastric fluid but exhibited low release in the intestinal fluid.	Rutz et al. (2016)
β-carotene	Yellow bell pepper (<i>Capsicum annuum</i>)	Ultrasonic homogenization, kneading	β-cyclodextrin	<ul style="list-style-type: none"> To prepare inclusion complexes using yellow bell pepper pigments and β-cyclodextrin by ultrasonic homogenization and kneading 	Isotonic beverage	Both processes resulted in good complex yield and inclusion efficiency.	Lobo et al. (2018)
Lycopene	Lycopene dispersed in oil (10% lycopene)	Spray Drying	Capsul Starch	<ul style="list-style-type: none"> • To determine the release profile of anthocyanins under simulated gastrointestinal conditions. • To evaluate the use of microparticles in pectin candy. 	Cake	Microencapsulation provided greater protection to lycopene in comparison with its free form.	Rocha et al. (2012)
Astaxanthin	Yeast (<i>Phaffia rhodozyma</i>)	Phase Separation	Zein and oligochitosan	<ul style="list-style-type: none"> To encapsulate astaxanthin by oligochitosan-zein complex formation 	Liquor, apple vinegar, and rice vinegar	Encapsulation effectively improved the UV-light and storage stabilities of the astaxanthin, exhibiting greater stability at 4 °C.	Jiang and Zhu (2019)

Table 3
Various extraction techniques to extract natural pigments.

Technique	Principle	Source	Solvent	Pigment	Yield (µg/mg sample)	Operating Condition	Advantages	Disadvantages	References
Ultrasound assisted extraction (UAE)	• Shock waves and microjets cause cellular matrix disruption. • Turbulence and acoustic streaming increase mass transfer.	<i>Bougainvillea glabra</i> flowers	50% methanol	Betacyanin Betaxanthin	1.72 5.78	Temperature: 55 °C, Time: 37 min, Power and frequency: 88 W, 20 kHz,	Fast extraction, lower extraction temperature, faster processing time	High cost, large solvent consumption, specialized setup required	(Jurić et al., 2020a; Ngamwonglumlert et al., 2017b)
Supercritical fluid extraction (SFE)	Extraction using a medium such as CO ₂ and variable pressure and temperature.	<i>Scenedesmus obliquus</i>	Solvent: CO ₂ with 7.7 mL/dL ethanol	Carotenoids Chlorophyll a Chlorophyll b Chlorophyll c	0.30 0.85 0.36 0.02	Temperature: 40 °C, Time: 240 min, Pressure: 250 bar,	Safe, fast, Moderate extraction time, No use of toxic solvents	Influenced by the property of the fluid used, Need of safety controls	Guedes et al. (2013)
Pressurized liquid extraction (PLE)	Using high pressure and higher temperatures to extract the compounds	Blackberry (<i>Rubus fruticosus</i> L.)	water with 50 mL/dL ethanol,	Anthocyanins	1.02	Temperature: 100 °C, Time: 30 min Pressure: 75 bar, Flow rate: 3.35 mL/min,	Faster extraction, higher operating temperature, rapid extraction, reduced solvent consumption	Extraction efficiency varies with temperature, pressure, and type of solvent used Not suitable for heat-sensitive pigments	(Machado, Pasquel-Reátegui, Barbero & Martínez, 2015)
Microwave assisted extraction (MAE)	Direct heating inside the matrix, a rapid increase of the local temperature and pressure	Carrots (<i>Daucus carota</i> var. <i>staivus</i>)	Solvent: mixed solvent (50 mL/dL hexane, 25 mL/dL acetone, 25 mL/dL ethanol),	Total carotenoids β-carotene	0.52 0.23	Temperature: 58 °C, Solvent volume: 75 mL, Microwave power: 180 W, Time: 3 min	Maximum anthocyanins yield obtained after 10 min using MAE	Energy consuming, External factors (temp., pressure, power frequency) influence the output and efficiency	Hiranvarachat, Devahastin, Chiewchan, and Vijaya Raghavan (2013)
Enzyme-assisted extraction	Disruption and degradation of cell walls, the release of bound target bioactive from macromolecule	Hexane	Marigold Flower (<i>Tagetes erecta</i>)	Carotenoids	0.5	Enzyme to sample ratio (mL/g): 0.1:1, Time: 60 min, Extraction temperature: 25 °C, Solvent to sample ratio (mL/g): 4:1,	Applicable for heat-sensitive pigments, Easy to handle Moderate solvent consumption	Filtration required, Non-automated system	Barzana et al. (2002)

developments in carotenoids particle production and coprecipitation with biodegradable polymers. The use of supercritical fluids, rapid expansion from gas antisolvent process (GAS), supercritical antisolvent (SAS), supercritical extraction from emulsions (SFEE), and particle from gas saturated solutions (PGSS) (Cocero, Martín, Mattea, & Varona, 2009). Because of the low temperatures of supercritical fluid, its non-toxicity, and its total removal from the end product, supercritical micronization is one of the current procedures for carotenoid encapsulation.

Carotenoids can be precipitated using supercritical fluid technologies, and the SAS method in particular can produce particles with mean diameters ranging from 1 to 200 µm (Mattea, Martín, & Cocero, 2009). The SAS method can be used to successfully coprecipitate microparticles of PEG loaded with bixin-rich extract. The SAS method can be used to successfully coprecipitate microparticles of PEG loaded with the bixin-rich extract (Santos & Meireles, 2013). The encapsulation of β-carotene was studied with two different polycaprolactones as coating agents by PGSS process (de Paz, Martín, Duarte, & Cocero, 2012).

Spray-drying, freeze-drying, and supercritical antisolvent precipitation of blackberry residue extracts resulted in particles with significant antioxidant capacity and little degradation of the target components. SAS precipitation of anthocyanins in polymeric matrix looks to be a promising technique for removing the extract's residual ethanol and

producing a dry product that is richer in anthocyanins and purer (Machado, Pasquel-Reátegui, Barbero, & Martínez, 2015). When compared to the thin film hydration approach, the supercritical carbon dioxide (SC- CO₂) method produced liposomes with improved intactness, sphericity, and uniformity. This technology allows for the processing of phospholipid aggregates into nano/micro particles using dense phase CO₂ and the control of their properties through the adjusting of processing parameters. The SC-CO₂ approach shows promise in the scalable manufacture of liposomes containing a wide range of bioactive for food applications (Zhao & Temelli, 2017).

8. Food applications of encapsulated pigments

In recent years, there has been an increasing trend toward using natural pigments as alternatives to synthetic colors in food applications, owing to both legal acts and consumer concerns about health. Encapsulation appears to be an effective method for protecting and stabilizing nanoparticles. Accessible literature data on the use of encapsulated natural pigments as coloring agents in food products is still rare, especially when compared to a large number of papers on encapsulation methods and production procedures of pigment microparticles and their in vitro characterisation. However, a better understanding of the issues surrounding the incorporation of encapsulated natural pigments into

various food matrices, as well as the complexities associated with the heterogeneous and composite formulation and structure of food, is required to enable the prediction of natural pigments behavior in real foods, in terms of both coloring capability and potential health benefits (Jurić et al., 2020). The use of plant pigments as natural food colorants exhibits a positive effect on the consumer demand for natural products and encapsulation is a method that can potentially protect the pigments from degradation. Application of the encapsulated pigments in the food industry is a new horizon in the development of functional food products. Lycopene microcapsules were produced by spray drying technique and modified starch was used as a coating material. These microcapsules were applied to the cake and they were capable to release the pigment during the preparation of the food system. The homogenous distribution of the color was observed in the sample with microencapsulated lycopene (Rocha, Fávaro-Trindade, & Grosso, 2012). Microparticles containing anthocyanin extract were prepared by the ionic gelation technique for the estimation of anthocyanins release profile under the simulated gastrointestinal environment and the use of microparticles in the jelly candy matrix. Jelly candy with particles produced by ionic gelation with dripping-extrusion technique exhibit greater retention of the bioactive compound in comparison with the atomization method. Palm oil and β -carotene were encapsulated with chitosan/sodium tripolyphosphate or chitosan/carboxymethyl cellulose. The microparticles encapsulated with chitosan/carboxymethylcellulose yield 87% whereas that of microparticles encapsulated with chitosan/sodium tripolyphosphate was 55%. The properties of these microparticles in food models were also determined. During application in the food system, these microparticles displayed improved carotenoid release but exhibited decreased carotenoids release during storage (Rutz, Borges, Zambiasi, Cleonice, & Médelin, 2016). Lycopene microparticles were produced by complex coacervation and freeze-drying and whey protein isolate and acacia gum were utilized as wall materials. After the storage at 4 °C for 14 days, the retention of lycopene was 63%. The powder was utilized for functionalization in dressing samples, exhibiting enhanced antioxidant activity (Gheonea (Dima) et al., 2020). The anthocyanins from sour cherries' skins extract were microencapsulated by using the freeze-drying method and whey protein isolates and acacia gum were used as coating agents. The encapsulated sour cherry extract is an excellent source of bioactive constituents and also can be utilized as a growth factor by the probiotic strain L. Casei. Both the extract and the powder exhibited good antioxidant properties and therefore can be used as functional component foods (Oancea et al., 2018). Despite the potential health-promoting properties, the use of anthocyanins in food products as a coloring agent is a difficult task because of its low stability during the processing and storage period. Encapsulation is an effective route to provide coating against detrimental conditions such as oxygen, humidity, and light (Yousuf, Gul, Wani, & Singh, 2016). The encapsulated anthocyanin pigment was utilized as an alternative to artificial color in jelly powder. A jelly with 7% encapsulated color exhibits higher scores for all sensory attributes and physicochemical evaluations in comparison with synthetic colorants (Akhavan Mahdavi, Jafari, Assadpour, & Ghorbani, 2016). β -carotene-loaded solid lipid microparticles (SLMs) stabilized with hydrolyzed soy protein isolate were incorporated in the yogurt. This method designated that it is an excellent technique with good potential use that can be applied in nutritional fortification or as the substitution of synthetic colorants in dairy products (Molina, Lima, Moraes, & Pinho, 2019). β -carotene-loaded solid lipid microparticles (BCSLMs) were incorporate into vanilla ice creams. The BCSLMs were prepared with palm stearin as the lipid phase, hydrolyzed soy protein isolate as the surfactant, and xanthan gum was used as the thickener. The incorporation of BCSLM into ice creams is a promising substitute to decrease the application of synthetic coloring agents to the products (Lima, Brito-Oliveira, & Pinho, 2016). β -carotene-loaded liposomes were produced using proliposomes and these were incorporated in yogurts. Liposome dispersions were stabilized by the mixture of xanthan and guar gums which was effective to avoid the aggregation of

β -carotene-loaded liposome during the period of storage. The liposomes were highly capable of protecting the encapsulated β -carotene from degradation for a storage time of 95 days. It is a feasible technique to produce yogurt with β -carotene encapsulated in liposomes as a functional component (Tonizazzo et al., 2014). To overcome the problems of low bioaccessibility and low stability, the betanin was loaded in liposomal nanocarriers which are significantly favorable owing to a) their potentiality to encapsulate both hydrophobic and hydrophilic constituents b) their amphiphilicity, non-immunogenicity, biocompatibility, and non-toxicity. After 60-days of storage, liposomal gummy candies exhibited significantly higher stability and antioxidant activity of betanin than gummy candies containing free betanin (Amjadi, Ghorbani, Hamishehkar, & Roufegarnejad, 2018). A study was conducted to assess the anthocyanin stability, encapsulated under various environmental conditions with various encapsulating materials in an isotonic soft drink system. The use of maltodextrin and gum arabic resulted in the prolonged anthocyanin half-life time, lowest degradation constant, and greater protection of the anthocyanin pigments (Burin, Rossa, Ferreira-lima, Hillmann, & Boirdignon-luiz, 2011). *Opuntia stricta* fruit juice is an important source of betacyanin pigments that can be applied as a natural red-purple food colorant. Spray drying is considered to be a vital method to produce a red-purple powder food colorant from *Opuntia stricta* fruit juice that exhibits high color strength with a drying yield of 58%. This powder colorant was applied to yogurt and a soft drink to develop foods of vivid red-purple tonalities (Obón, Castellar, Alacid, & Fernández-López, 2009). Betaxanthins from cactus pear fruits is a propitious natural yellow colorant in the cases of low-temperature storage and non-transparent packaged foods. A betaxanthin-rich water-soluble food colorant was prepared by spray-drying technique by utilizing maltodextrin as a coating agent. The use of the colorant microcapsules was successfully evaluated in yogurt and a soft-drink and there was excellent preservation after 28 days at 4 °C in both model systems (Fernández-lópez, Roca, Angosto, & Obón, 2018). Betalain-rich capsules were produced by ionic gelation with calcium alginate from a betalain-rich extract. Gummy candies were incorporated with these capsules and the texture characteristics and colorimetric stability of this food system were evaluated. These gummy candies exhibited excellent gelling and morphological characteristics that can be applied in confectionery industries (Otálora, de Jesús Barbosa, Perilla, Osorio, & Nazareno, 2019). A study regarding encapsulation of astaxanthin with zein and oligochitosan enhanced its storage stabilities and improved its application in the food industry significantly (Jiang & Zhu, 2019).

9. Conclusion and future perspectives

Color is an essential attribute of food that contributes to the evaluation of food quality. Pigments are intriguing functional components that are currently getting a lot of attention from researchers and consumers due to their safety, as well as their nutraceutical properties and biological activity. Apart from their importance as natural colorants, they are considered as health promotional ingredients that exhibit several beneficial functions and are a promising alternative to synthetic colorants. Pigments have a variety of properties that limit their use in food, including sensitivity to environmental and process stresses, low bioavailability, and low water solubility. Natural pigments respond to rising consumer demand for healthier, more natural foods to eventually replacing synthetic colorants entirely. Multiple factors have hampered the commercial development of natural pigments to date, including the limited range of natural colors approved for food use and the time-consuming process for regulatory approval of novel colourants, the higher cost in comparison to synthetic colourants, and the large volumes of biomass required for extraction per unit mass of natural colorant.

Future technological challenges are mainly induced by the exploration of enhancing the color intensity and stability of natural pigments by microencapsulation and nanoencapsulation techniques, and designing pigment mixtures to strengthen the properties of the desired food

product. The encapsulation method has various applications for encapsulating colorants, not only in the food industry, but also in the textile, pharmaceutical, and other industries. In the food sector, encapsulation of pigments serves a variety of purposes, including disguising undesired flavour or taste, preserving unstable elements, incorporating extra functional and nutritional components, and site-specific, controlled release of encapsulated pigments. Current studies are displaying new steps concerning the diverseness of colorants that are applied to substitute synthetic colorants with natural ones. Natural pigments in functional foods can now be valorized by encapsulation, bringing their benefits to a wider population. Microencapsulation and nanoencapsulation are effective platforms for protecting pigments from harmful environmental conditions while also allowing for controlled and targeted release. The application of micro and nano-encapsulates in various aspects of food and gastrointestinal systems must be investigated to explain their release mechanisms and application efficiency. Future natural pigments research should also focus on expanding the range of colors that may be obtained, as well as encouraging pigments with health-beneficial qualities. More research is also needed on the stabilization of natural colorants, which has so far been addressed via molecular complexation or microencapsulation techniques. (Figs. 1–4), (Tables 1–3)

In the future, more research is needed in the following areas:

- ✓ How to improve the absorption efficiency of encapsulated pigments.
- ✓ Which encapsulating agent is more responsible for the bio-accessibility and bioavailability of the encapsulated pigments.
- ✓ Benefits of encapsulated pigments on human health.
- ✓ Identifying innovative encapsulation methods and assessing the efficacy of their use in food or biological systems.
- ✓ Further research into more efficient production processes to improve extraction yield and reduce pigment extraction time.
- ✓ Developing new approaches for the stabilization of natural plant pigments to widen their application in the food industry.

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