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Stability Enhancement of Natural Food Colorants- A Review

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Abstract
The public knowledge about the importance of using natural ingredients in food products opened new areas toward the extraction, stabilization, storage, and application of natural colors. The stabilization of natural colorants has a key role in their application in food industries. Colorants are significantly sensitive out of their natural resources and some of them start to fade rapidly after extraction. In this paper, the most applicable methods for the stabilization of natural colorants in the food industry were reviewed. The present paper aims to review the published scientific researches about stabilization methods of different natural colorants. The Google Scholar, PubMed, Web of Science databases were searched. Among 120 final selected papers, 73 related papers have used. The review starts from the manuscripts published in 2020 and then continued to review the manuscripts published in 2000. Different kinds of literature reported stabilization methods of major natural colorants like anthocyanins, carotenoids, chlorophylls, and betalains. These ways can be applied to food studies before, during, and after the formulation and manufacturing of food products. Because of the different roles of natural colorants in human health and also the oxidative stability of foods, it may be a good choice to stabilize and application of natural colorants in food products.

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Introduction
Despite the coloring and nutraceutical potential of natural colorants, there are some limitations to incorporate them into food and beverage formulations. Sometimes the type of colorant as a hydrophobic or hydrophilic molecule, the hygroscopic nature (Weigel, Weiss, Decker, & McClements, 2018), the stability to the chemical degradation (due to oxidation) (Cortez, Luna-Vital, Margulis, & Gonzalez de Mejia, 2017), high processing temperature and pressure (Selig *et al.*, 2020), light, acidity and certain components in the food formulation, and stability of natural colorant during storage, makes them hard to use in food and beverage products.

Colors are used in the food industry to retain, increase the intensity of the natural color of food, or to make a new hue in food products (Natália Martins, Roriz, Morales, Barros, & Ferreira, 2016; Natalia Martins, Roriz, Morales, Barros, & Ferreira, 2017; Mojica, Berhow, & Gonzalez de Mejia, 2017; Torres *et al.*, 2016; Weber, Boch, & Schieber, 2017). Behind the coloring properties, there are different health benefits from natural colorants (Bandeira *et al.*, 2017; Campos *et al.*, 2017).

Natural colorants can classified based on their source, their solubility in water or oil, and their chemical structure. The most common way for classification of natural colors are based on their chemical structure. The carotenoids, anthocyanins, betanins, and chlorophylls are the most natural colors used in food industries (Rodriguez-Amaya, 2016).

Some pigments like anthocyanins have a wide range of colors and also have antioxidant and potential health benefits that make them a good choice for coloring food products (Weber *et al*., 2017; Zhao *et al*., 2017). Some like lycopene have an orange-red color, antioxidant, antibacterial (Ranjbar & Ranjbar, 2016), and also health benefits that make them suitable to use as well as a functional ingredient (Ranjbar Nedamani, Ranjbar Nedamani, & Salimi, 2019; Sroynak, Srikalong, & Raviyan, 2013; Sultan Alvi, Ansari, Khan, Iqbal, & Khan, 2017; Zhang *et al*., 2016).

Todays because of the growing demand for natural ingredients in food products, and “clean” labeling considerations, the food industries are trying to applicate natural colorants, flavorings, and other ingredients to their food products. Natural colorants have a wide range of applicability in the food industry. Colors like anthocyanin, β-carotene, lycopene, betalain, chlorophyll, riboflavin, curcumin, and so on are used in different foods and beverages (Calvo & Salvador, 2000; Chung, Rojanasasithara, Mutilangi, & McClements, 2017; Cortez *et al*., 2017; Francis & Markakis, 1989; Leong, Show, Lim, Ooi, & Ling, 2017; Rodriguez-Saona, Giusti, & Wrolstad, 1999; Sigurdson, Tang, & Giusti, 2017; Yin, Fei, & Wang, 2017; Yusuf, Shabbir, & Mohammad, 2017). But the most challenging issue of using natural colorants in food products is their stability during and after processing. Also behind the color stability problems (compared with synthetic colors), some doubt exists about the amount of natural colorant absorption in the human body after consuming a nutraceutical or functional food containing natural colorants. Thus different methods are used for the stabilization of natural colorants. Some methods like emulsion-based delivery systems (Weigel *et al.*, 2018), acylation (Zhao *et al.*, 2017), encapsulation (microencapsulation and nanoencapsulation), copigmentation (Weber *et al.*, 2017), complex-forming with polymers, peptides, and amino acids (Chung, Rojanasasithara, Mutilangi, & McClements, 2015; Chung *et al.*, 2017), and metal-complexation is used to protect and deliver natural colorants to improve their application in food and beverage formulations. The application of each stabilization method depends on the conditions that food is produced, handled, and stored (Weigel *et al*., 2018). Also, some factors like the nature of colorants, light exposure, and the oxidation factors should be undertaken (Delgado-Vargas, Jiménez, & Paredes-López, 2000; Espín, Soler-Rivas, Wichers, & García-Viguera, 2000; Francis & Markakis, 1989).

In this paper, we have reviewed the stabilization methods of natural colorant for use in food products. This review is based on methods that are most studied on different natural colorants. Between them may be more than one method is used for anthocyanin's stabilization. Perhaps because of (1) the extent of studied which more of them is about different methods of anthocyanin stabilizations (Bassa & Francis, 1987; Bastos, Oliveira, Melo, & Lima, 2017; Giusti & Wrolstad, 2003; Mojica *et al.*, 2017; Rodriguez-Saona *et al.*, 1999) (due to (2) their high sensitivity to different factors like pH, temperature, other components in food formulation, oxygen, and so on), (3) the wide range of colors of anthocyanin that make them suitable for use in food products, and (4) that some colorants like carotenoids are mostly stabilized by encapsulation methods (Chung, Rojanasasithara, Mutilangi, & McClements, 2016b; Natália Martins et al., 2016).

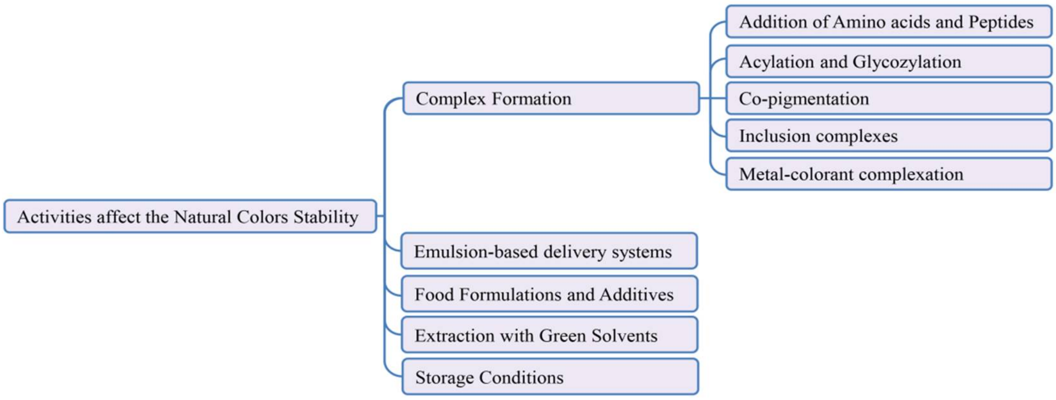

Fig. 1. A summarize of stabilization methods for natural food colorants in this review

However, this literature review aimed to make a classified paper about different stabilization methods for different natural colorants (Fig. 1). Also, review the most and the least colorant stabilization studies to make a good viewpoint for the last stabilization studies of natural colorants. Some colorants with new natural sources or extraction methods are studied that can be good choices to study their stability in food production or their stabilization methods.

Stabilization methods of natural pigments

The natural colorants are stabilized through different methods. The selection of stabilization methods depends firstly on the nature and the structure of the colorant (Chung *et al.*, 2017; Leong *et al.*, 2017; Ludin *et al.*, 2014; Selig *et al.*, 2020; Zhao *et al.*, 2017). The mechanism in wich the natural color losses, can also be a good way for choosing the suitable stabilization method. Fig. (2) shows some natural colorant degradation mechanisms. According to the literature some factors such as pigment concentration, degree of glucosylation and acylation, water activity, presence of degrading enzymes such as polyphenol oxidize, metal ions, pH, temperature, light, oxygen, and water concentration highly affect natural colorant stability (Cortez *et al.*, 2017; Delgado-Vargas & Paredes-López, 2003; Huang *et al.*, 2016; Khan, 2016; Lourith & Kanlayavattanakul, 2011; Selig *et al.*, 2020; Vendruscolo *et al.*, 2013; Weber *et al.*, 2017; Yin *et al.*, 2017). There are some types of natural colorant stabilization. Some are just suitable for anthocyanins and some are general and can be used for more colorants. The following sections are based on a literature review to make a classified view for natural colorants stabilization methods. Tables (1) summarized the stabilization methods according to colorant and scientific source by their obtained results:

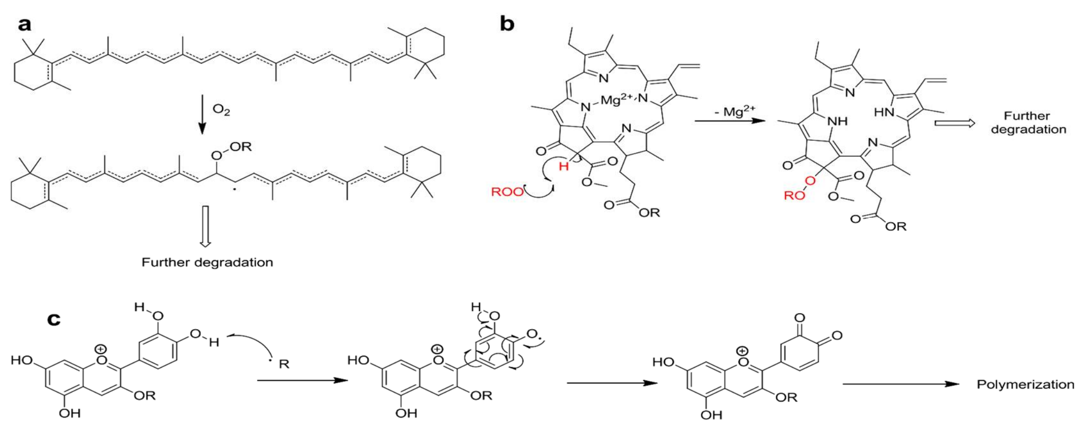

Fig. 2. Mechanisms of color loss: (a) β- carotene oxidation, (b) chlorophyll peroxidation and magnesium losing, (c) anthocyanin radical oxidation (Adapted from Ghidouche, Rey, Michel, & Galaffu (2013) with permission from Elsevier)

Table 1. Summarized literature review in natural colorant stabilization methods and their application in food

Type of colorant	Stabilization component	Stabilization method	Food model	Result	Reference
Anthocyanin	native whey protein; denatured whey protein; citrus pectin; and beet pectin	Addition of amino acids and Peptides	Beverage	Denatured can significantly enhance the stability of the anthocyanin due to hydrogen bonding between anthocyanin and whey protein	(Chung *et al*., 2015)
purple carrot anthocyanins	three amino acids (L-phenylalanine, L-tyrosine, L-tryptophan) and a polypeptide (e-poly-L-lysine)	Addition of Amino acids and Peptides	Model beverages	Amino acids or peptide (0.1%) can increase the color stability, especially for l-tryptophan	(Chung *et al*., 2017)
grape skin anthocyanin	preheated casein (at 40- 100°C) and whey proteins (at 45- 60°C) (for 15 minutes)	Addition of Amino acids and Peptides	Color loss and anthocyanin degradation of grape skin extract	1- Preheating of casein and whey proteins can increase the stability of grape skin anthocyanins during thermal treatment (at 80°c for 2h), oxidation with h_2o_2 (0.005% for 1h), and illumination (at 5000 lx for 5 days) 2- Preheated whey proteins have better effects on stability than preheated casein	(He, Xu, Zeng, Qin, & Chen, 2016)
B. boliviana anthocyanins	acylation	Addition of Amino acids and Peptides	Naturally colored yogurt	The half-life of nonacylated pigments was 125 and 104 day (at 10 and 20 mg cy-3-glu equivalents/100 g yogurt), and acylated pigments have 550.2, 232.6, and 128.9 d half-life (at 20 mg of cy-3-glu equivalents/100 g of 4%, 2%, and 0% fat yogurt)	(Wallace & Giusti, 2008)
corn anthocyanins		Copigmentation	Beverage model	1- Cyaniding-rich model beverage showed higher stability but flavone-rich extract increased the half-lives of both pigments. 2- The acylation had a weaker effect on half-lives	(Chatham, Howard, & Juvik, 2020)
Grape skin anthocyanins	Quercetagetin	Copigmentation		Increase in the half-life of color and most effective than epigallocatechin gallate, tea polyphenols, myricetin, and rutin	(Xu, Liu, Yan, Yuan, & Gao, 2015)
red bell pepper pigments	β-cyclodextrin	inclusion complex (through magnetic stirring and ultrasonic homogenization)	Yogurt	Ultrasonic homogenization has a better effect on color stability and the color indices of colored yogurt	(Gomes, Petito, Costa, Falcao, & de Lima Araujo, 2014)
chlorophyll extracts	Zinc and copper	Metal-colorant complexation	Against combined acid-heat conditions in formulated beverages	1- Metal complexation of chlorophyll increases the stability of green color 2- These complexes can make a higher hue color	(Ngamwonglumlert, Devahastin, & Chiewchan, 2017)

Table 1. Summarized literature review in natural colorant stabilization methods and their application in food

Type of colorant	Stabilization component	Stabilization method	Food model	Result	Reference
Cyaniding-3-glucose	Fe^{3+} and alginate	Metal-colorant complexation		1- A combination of metal and alginate has a graduate effect on color stability and intensity increasing 2- The effect depends on pH	(Tachibana, Kimura, & Ohno, 2014)
Cyanidin and delphinidin derivatives	Al^{3+} salt	Metal-colorant complexation		1- Salt ration, ph, and colorant concentration are critical in color intensity. 2- Chelating the al^{3+} by anthocyanins leads to producing a variety of intense violet to blue colors under acidic pH	(Gregory T Sigurdson & Giusti, 2014)
Lutein	B-lactoglobulin, β-lactoglobulin/lecithin, Biozate 1 (the whey protein hydrolysate), Biozate 1/lecithin, Tween 20, and Tween 20/Lecithin	Emulsion-based delivery system	Lutein uptake		(Frede *et al.*, 2014)
Betalain	Maltodextrin, guar gum, Arabic gum, pectin, and xanthan gum	Emulsion-based delivery system		Freeze-drying method and maltodextrin with xanthan and guar gum (as coating agent) increase the stability of betalain	(Ravichandran *et al.*, 2014)
Betalain red dye (extracted from *beta vulgaris* L. (beet))		Emulsion-based delivery system	Comparison with commercial dye FD&C Red 40	Encapsulated betalain red dye is more resistant to uv light. It had a high sensitivity to changes in pH and temperature	(Hernández-Martínez *et al.*, 2017)
B-carotene	A water-soluble mixture of Ca^{2+} cross-linked alginic acid	Emulsion-based delivery system		Calcium can increase the nanoparticle density and improve the colorant stability against the oxidation	(Astete, Sabliov, Watanabe, & Biris, 2009)
Color and anthocyanins	Preparation under nitrogen, CO_2, and air	Storage conditions	Strawberry puree		(Howard, Brownmiller, & Prior, 2014)
Betalain	Spray drying and freeze-drying for comparison to the effect of drying methods	Storage conditions		Freeze-drying method and maltodextrin with xanthan and guar gum (as coating agent) increase the stability of betalain	(Ravichandran *et al.*, 2014)

Table 1. Summarized literature review in natural colorant stabilization methods and their application in food

Type of colorant	Stabilization component	Stabilization method	Food model	Result	Reference
Roselle extract	Freeze-dried	Storage conditions	Beverage model system	Lightness and hue changes were low during storage	(Duangmal, Saicheua, & Sueeprasan, 2008)
C-phycocyanin (c-pc) (a blue pigment often found in cyanobacteria)	Citric acid addition	Food Formulations and Additives	Incubation at 80 °c for 1h.	During processing under high temperature and extraction, citric acid can increase the stability of c-phycocyanin	(Pan-utai, Kahapana, & Iamtham, 2017)
C-phycocyanin	Natural protein crosslinker methylglyoxal, honey, and a high concentration of sugar	Food Formulations and Additives		Methylglyoxal does not a significant effect on color stabilization. Honey and a high concentration of sugar can play a preservative role in color stabilization. They believed that the preservative effect of high concentration of sugar on the blue color of c-phycocyanin is dependent mostly on the final concentration of sugar in solution than the sugar type	(Martelli, Folli, Visai, Daglia, & Ferrari, 2014)
Betaxanthins	Ph and temperature adjustment, gum	Food Formulations and Additives	Beverage	At ph 6.6 and the presence of ascorbic acid as an additive, the maximum stability of betaxanthins achieved	(Rodriguez-Sanchez, Cruz, & Barragan-Huerta, 2017)
Annatto (orange), chlorophyllins (green), cochineal (red), and curcumin (yellow)	Gelatin, a mixture of xanthan and locust bean gum, sugar, and natural colorant	Food Formulations and Additives	Gel	Cochineal and curcumin natural colorants can replace the synthetic ones in jellies	(Calvo & Salvador, 2000)
Roselle extract	Maltodextrin and trehalose as a stabilizer	Food Formulations and Additives	Beverage model system	Lightness and hue changes were low during storage. Maltodextrin had a higher stabilizing effect than trehalose	(Duangmal *et al.*, 2008)

Complex formation

Addition of amino acids and peptides

Complex formation between natural colorants and non-color molecules (for example co-pigments) can improve their stability, especially in food products. Polysaccharides, peptides, pectin, biopolymers, and phenols that are used in food formulations, can be applied to form a stabilized complex (Chung *et al.*, 2015). Chung *et al.* (2015) studied the addition of the %1 native whey protein effect; denatured whey protein; citrus pectin; and beet pectin in beverage formulation containing anthocyanin (0.025%), ascorbic acid (0 or 0.05%), and calcium salt (0 or 0.01%) on anthocyanin stability under accelerated conditions at 40 °C for 0 to 7 days with exposure to ambient light (Chung *et al.*, 2015). They found biopolymers, particularly whey protein which is denatured with heat, can significantly enhance the anthocyanin stability due to hydrogen bonding between anthocyanin and whey protein (Chung *et al.*, 2015). They also studied the role of three amino acids (L-phenylalanine, L-tyrosine, and L-tryptophan) and a polypeptide (e-poly-L-lysine) in model beverages on purple carrot anthocyanins stability at elevated temperature (40 °C/7 days) (Chung *et al.*, 2017). They found amino acids or peptides (0.1%) can increase the stability of color, especially for L-tryptophan from 2-6 days (Chung *et al.*, 2017). Yi, Fan, Yokoyama, Zhang, & Zhao (2016) also investigated the effect of complex formation between lutein and whey protein isolate (WPI) and sodium caseinate (SC) on hydrophobic lutein stability during 16 days storage (Yi *et al.*, 2016). They found the complex formation of lutein can increase stability and this stability increased with an increase in protein concentration. He *et al.* (2016) studied the effect of preheated casein (at 40-100 °C) and whey proteins (at 45-60 °C) (for 15 min) on color loss and anthocyanin degradation of grape skin extract at pH 3.2 and 6.3 (He *et al.*, 2016). They found preheating of casein and whey proteins can increase the grape skin anthocyanins stability during thermal treatment (at 80 °C for 2 h), oxidation with H_2O_2 (0.005% for 1 h), and illumination (at 5000 lx for 5 days). Also, the preheated whey proteins have better effects on stability than preheated casein (He *et al.*, 2016). The results of studies like He *et al.* (2016) can be important from the nutritional viewpoint either from that stabilization of a nutraceutical colorant and application of milk proteins in food products as a carrier or as a stabilizer for natural ingredients. Casein and whey proteins have widespread use in the food industry because they have nutritional and functional properties in food products.

Acylation and glycosylation

Because of the chemical instability of anthocyanins, stabilization is an important way to make them suitable for food production. The most abundant anthocyanidins that are used in food formulations are cyanidin, delphinidin, and pelargonidin, and also malvidin, petunidin, and peonidin (Bastos *et al.*, 2017; Caldas-Cueva *et al.*, 2016; He *et al.*, 2016; Rodriguez-Amaya, 2016). Different factors like temperature, light, and presence of other phenolic compounds, metal ions, vitamin C (Chung *et al.*, 2017; Xu *et al.*, 2015), and oxygen can affect the anthocyanins stabilization (Qian, Liu, Zhao, Cai, & Jing, 2017) (Fig. 3). But pH and temperature have a significant effect on anthocyanin stability (Luna-Vital, Li, West, West, & Gonzalez de Mejia, 2017).

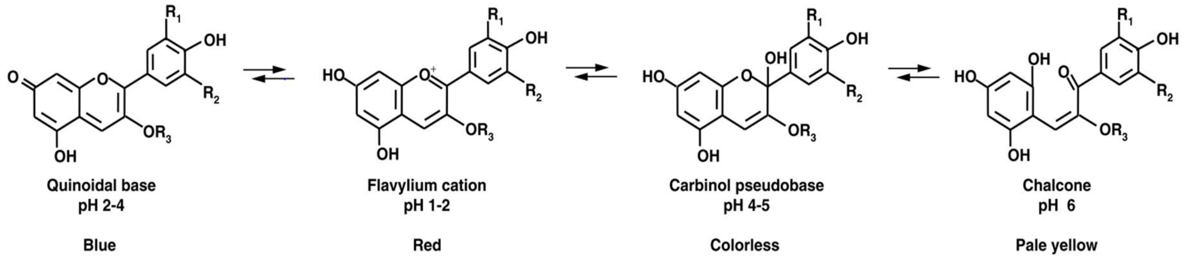

Fig. 3. Anthocyanins structure and color changes under different pH (Adapted from Rodriguez-Amaya (2016) with permission from Elsevier)

The stability of anthocyanins increases with glycosylation and acylation methods. Cortez *et al.* (2017) believed that the chemical stability of acylated anthocyanins increases with glycosyl acylation in vitro and in vivo. Most anthocyanins are naturally (in plant vacuoles) in their acylated forms (Zhao *et al.*, 2017). Glycosyl acylation has an important role in the stabilization of the anthocyanins and makes different types of them (Zhao *et al.*, 2017).

The anthocyanin can bind with glycosylating groups, such as galactose, xylose, arabinose, and sugars such as glucose, and rhamnose, through hydroxyl groups of aglycone part and produce glycosidic bonds. Then it can further link through ester bonds to the sugars. Also, it can acylate with organic acids such as cinnamic acids or aliphatic acids. In anthocyanin glycosyl acylation (anthocyanin acylation), the -OH groups of anthocyanin glycosyls partially esterified with organic acids such as acetic acid, oxalic acid, L-lactic acid, malonic acid, succinic acid, malic acid, tartaric acid, glutaric acid, erucic acid, p-hydroxybenzoic acid, gallic acid, (E)-p-coumaric acid, (E)-caffeic acid, (E)-3,5-hydroxycinnamic acid, (E)-ferulic acid, (E)-sinapic acid. This phenomenon is catalyzed by acyltransferases in cytosol and vacuoles (Zhao *et al.*, 2017).

This glycosylation (intramolecular H-bonding network within the sugar moiety, between the sugar moiety and the anthocyanin molecule) or acylation processes can change the molecular size and polarity of anthocyanin. Higher glycosylation can increase the polarity and water solubility, and higher acylation can reduce polarity and water solubility. The type of sugar or acyl substituents in anthocyanins, make a wide variety of derivatives in nature.

Wallace & Giusti (2008) reported that acylation of the natural pigment of *B. boliviana* anthocyanins increases the stability of colorant in naturally colored yogurt. They show the half-life of nonacylated pigments was 125 and 104 days (at 10 and 20 mg cy-3-glu equivalents/100 g yogurt), and acylated pigments have 550.2, 232.6, and 128.9 d half-life (at 20 mg of cy-3-glu equivalents/100 g of 4, 2, and 0% fat yogurt) (Wallace & Giusti, 2008).

Copigmentation

Copigmentation is a non-covalent interaction to form a complex of anthocyanins with a copigment (Chatham *et al.*, 2020; Delgado-Vargas & Paredes-López, 2003). Co-pigments are rutin, cinnamic acids, and ferulic acid (Weber *et al.*, 2017), pectin, and whey proteins, gums, cyclodextrins, phenolic compounds (Chatham *et al.*, 2020; Cortez *et al.*, 2017), metal ions, colorless compounds (Chung, Rojanasasithara, Mutilangi, & McClements, 2016a). Copigmentation has optical effects on anthocyanins color such as (1) hyperchromic shifts which leads to absorbance increase and has a darkening effect on color, (2) bathochromic shift which leads to increase the wavelength to maximum absorbance and can contribute to making a bluish-purple color (Chatham *et al.*, 2020). Copigmentation is a color shift (bathochromic shift) at a visible wavelength from red to blue (this phenomenon is called the blue effect). This is a process to increase the intensity of anthocyanin color (hyperchromic shift) (Bechtold & Mussak, 2009). In the plant, copigmentation of anthocyanin with phenolic compounds have a key role in color stabilization (Xu *et al*., 2015). At higher concentrations of pigments, the bluing effect of copigmentation is generally observed, and at low concentrations the yellowing effect is observed (Delgado-Vargas & Paredes-López, 2003).

Generally, the magnitude depends on hydrogen bond donor and acceptor, and the extension of π-conjugation involved flavones and flavonols of strongest co-pigments (Chatham *et al*., 2020). Copigmentation also increases the stability of colors by increasing the pK_a and provides a pH range that the flavylium cation is predominant in. This leads most anthocyanin molecules to remain in the colored flavylium ion forms despite the increase in pH (Chatham *et al*., 2020). This protective effect of copigmentaion can also increase the chemical stability of color (Chatham *et al*., 2020). Chatham *et al*. (2020) investigated the application of co-pigmented corn anthocyanins in a beverage model. They described C-glycosyl flavone and anthocyanin copigmentation system consists of pelargonidin and cyaniding-derived anthocyanins. The found cyaniding-rich model beverage showed higher stability but flavon-rich extract increased the half-lives of both pigments. They also reported that the acylation had a weaker effect on half-lives (Chatham *et al*., 2020).

Copigmentation can prevent the flavylium moiety hydration and thus stabilize it to make a red color in acidic conditions (Weber *et al*., 2017). Then the anthocyanin can be dried in its stable color. The drying factors and method are important to eliminate the moisture from the feed. The final moisture is critical in the hydration rate of co-pigmented color.

The copigmentation process is highly dependent on the pigments and co-pigments concentration (Qian *et al*., 2017). The type of co-pigment (such as phenols, flavanol, chlorogenic acid, caffeic acid, or rutin and polyphenols, such as tannins) also has a key effect on final stability and shelf life. The pH has a great role in co-pigmentation; copigmentation is weak at pH lower than 2 compared to pH= 2-5 (Bechtold & Mussak, 2009). Rutin and ferulic acid besides the copigmentation effect, have also antioxidative properties that prevent oxidation after drying and thus increase the color shelf life (Weber *et al*., 2017).

Copigmentation is one of the acylation methods of anthocyanins. It can protect the chromophore of the colorant from the nucleophilic attack of water at position 2 of the pyrylium nucleus and thus stabilize the color (Qian *et al*., 2017). Copigmentation is a natural and valuable procedure to stabilize anthocyanins (Bechtold & Mussak, 2009) (Fig. 4).

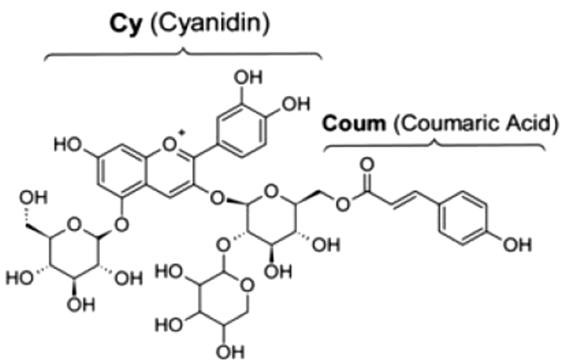

Fig. 4. Co-pigmented anthocyanin with coumaric acid (Adapted from Silva, Freitas, Maçanita, & Quina (2016) with permission from John Wiley and Sons)

Depending on the type of anthocyanin and co-pigments, copigmentation can make complexes through various mechanisms like self-association, intramolecular co-pigmentation, intermolecular copigmentation (Fig. 5), and metal complexation (Chung *et al*., 2016a). The self-association of anthocyanin stabilize molecule through copigmentation process (Qian *et al*., 2017).

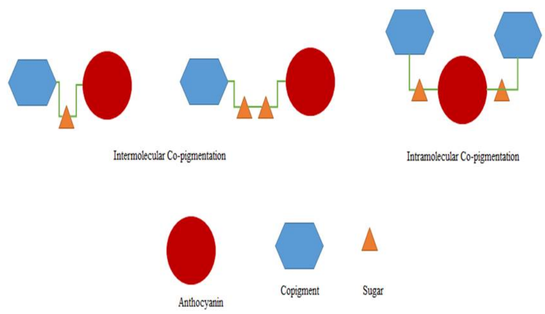

Fig. 5. Anthocyanin interactions (Adapted from Bechtold & Mussak (2009) with permission from John Wiley and Sons)

Intermolecular copigmentation is an interaction between anthocyanins and non-color co-pigment with noncovalent binding (such as hydrogen bonding, hydrophobic interactions, and electrostatic interactions) to the anthocyanin. Intermolecular copigmentation involved nonacylated anthocyanins in fruits (Bechtold & Mussak, 2009). However, intramolecular copigmentation due to covalent bonds are stronger than intermolecular copigmentation and can be more effective in stabilizing the color. This type of copigmentation can be found in the flower and vegetable acylated anthocyanins (Bechtold & Mussak, 2009). Self-association is a stacking-like interaction that takes place during wine aging (Bechtold & Mussak, 2009). This leads to forming the anthocyanin polymers although the condensation of heavy polymers of co-pigmented anthocyanins may result in the loss of color if double bonds are disrupted (Kaimainen, 2014).

Xu *et al.* (2015) studied the stability of grape skin anthocyanins copigmented with quercetagetin. Quercetagetin is structurally similar to quercetin and found to be the most copigment in diffused *Tagetes erecta* L. (marigold) flower. Quercetagetin also has antioxidative, anti-enzyme, and anti-microbial activity and can control diabetes type II. Thus Xu *et al.* (2015) used the pH values of 3, 4, and 5, the molar ratio of 1:10, 1:20, and 1:40 of anthocyanin: quercetagetin, at temperatures of 70, 80, and 90 °C with 5:1, 1:1, 1:5, and 1:10 light exposure ration experiments. They found the half-time of grape skin anthocyanin was significantly increased by quercetagetin concentration increasing and quercetagetin was most effective copigment than epigallocatechin gallate, tea polyphenols, myricetin, and rutin for stabilizing the grape skin anthocyanin (Xu *et al.*, 2015).

Inclusion complexes

The inclusion of bioactive components in the cavity of biopolymers like cyclodextrins can increase their stability, solubility, and bioavailability (Gomes *et al.*, 2014). Cyclodextrins have a cyclic structure contains 6, 7, and 8 D-glucose units (named as α-cyclodextrin, β-cyclodextrin, and γ-cyclodextrin, respectively). Their surface is hydrophilic and the cavity inside them is hydrophobic (Fig. 6); this leads them to be able to make stable inclusion complexes with drugs, colors, flavors, and other food or bioactive components (Gomes *et al.*, 2014). Cyclodextrins can be used to form complexes with natural lipophilic colorants to make them soluble in aqueous media. Since the β-cyclodextrin is more acceable and has low-cost than the other types of cyclodexterins, generally used for these purpose (Del Valle, 2004).

Methods such as co-precipitation, slurry complexation, paste complexation dampmixing and heating, extrusion, and drying mixing can be used for inclusion in cyclodexterins. After the process, the complexes dried trough oven, fluidized bed dryers or other methods (Del Valle, 2004).

Gomes *et al.* (2014) prepared an inclusion complex with red bell pepper pigments and β-cyclodextrin (mass ratio 1:4) to use in yogurt (Gomes *et al.*, 2014). They used two different procedures to make the inclusion complex: magnetic stirring and ultrasonic homogenization. They found ultrasonic homogenization has a better effect on color stability and the color indices of colored yogurt are better than the magnetic stirring complexes (Gomes *et al.*, 2014).

Because the cyclodextrins can form solid inclusion complexes (named host molecule) with solid, liquid, and gaseous compounds (named guest molecule), their application in color stabilization by this method is considerable. The dimensional fit between host and guest molecules is necessary. In this method, the non-polar moiety that has an appropriate size can enter into the lipophilic cavity of the cyclodextrin molecule and form an inclusion complex by non-covalent bonds (Del Valle, 2004).

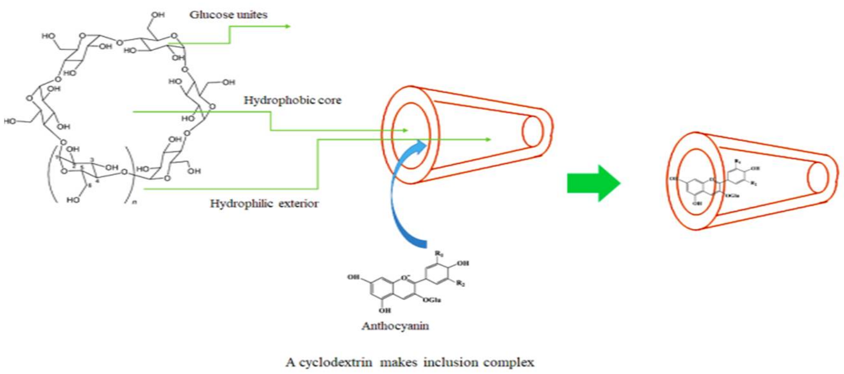

Fig. 6. Inclusion of anthocyanin in a cyclodextrin cavity

During the complex formation, the enthalpy-rich water molecules release from the cyclodextrin cavity and make the driving force of complex formation. Then the more hydrophobic guest molecules of solution enter into the cavity and make the cyclodextrin ring form a stable state with lower energy (Del Valle, 2004). The binding power between host and guest depends on the fitness between them and some specific interactions take place in surface atoms. Both in solution, or crystalline form complex can be formed but the water is dominant.

Metal-colorant complexation

Sometimes the addition of metals such as tin, iron, aluminum, magnesium, potassium to the colorant structure can stable or recolor the chromophore. When a colorant has more than one free hydroxyl group in its chromophore, the chance of metal-chelating increases (Cortez *et al.*, 2017).

The best example is chlorophyll which is a sensitive colorant to enzymes (Mg-dechelatase, chlorophyllase, and other oxidative enzymes such as lipoxygenase, chlorophyll oxidase, and peroxidase (Ngamwonglumlert *et al.*, 2017)), heat, and acidity. Blanching can destroy or decrease these enzymes' activity. But it makes an acidic condition that is not suitable to stabilize the green color of chlorophyll.

Metal complexes of chlorophyll such as copper- or zinc-chlorophyll are widely used to make a green color in food formulations. Insertion of copper or zinc is done to be replaced with a magnesium atom of chlorophyll structure. This process leads to the stabilization of the green color of chlorophyll. Ngamwonglumlert *et al.* (2017) studied the stability of the molecular structure, and zinc- and copper-chlorophylls extracts cytotoxicity against combined acid and heat conditions (Ngamwonglumlert *et al.*, 2017). They found metal complexation of chlorophyll increases the stability of green color and these complexes can make a higher hue color in formulated beverages. Also, they found zinc- and copper-chlorophylls have slightly higher cytotoxicity than untreated/steamed leaves and synthetic colorants (Ngamwonglumlert *et al.*, 2017). Tachibana *et al.* (2014) studied the effect of metal cation and polysaccharides addition on the natural stability in the acidic range. They use Fe^{3+} and alginate and investigate the cyaniding-3-glucose (C3G) stability at different pH and temperatures. They found the Fe^{3+} increases the color intensity after 50 min incubation but suddenly the color reduces due to the formation of aggregates in solution. Also, the alginate cannot have any effect on color by itself. But when a combination of Fe^{3+} and alginate polysaccharide was used, the color intensity and stability increased gradually. Also, this stability is dependent on pH (Tachibana *et al.*, 2014). Sigurdson & Giusti (2014) studied the effect of Al^{3+} salt on Cyanidin and delphinidin derivatives stability at pH 3-6 during 28 days. They found salt ration, pH, and colorant concentration is critical in color intensity. Also, chelating the Al^{3+} by anthocyanins under acidic pH, leads to producing a variety of intense violet to blue colors (Sigurdson & Giusti, 2014).

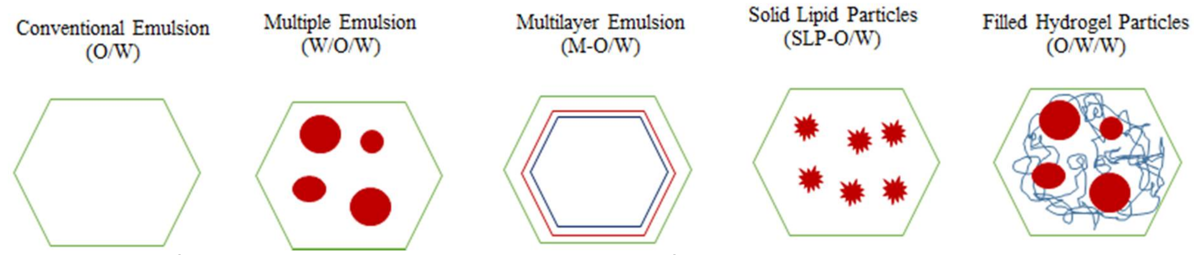

Fig. 7. Types of emulsion-based delivery systems (Adapted from McClements, Decker, & Weiss (2007) with permission from John Wiley and Sons)

Emulsion-based delivery systems

The most common methods of the emulsion-based system of color stabilization are oil in water systems (r>100 nm) and nanoemulsions (r<100 nm) (Weigel *et al.*, 2018) (Fig. 7). These systems can be used to improve water dispersion, increase the bioaccessibility and stability of bioactive components to chemical changes. When a natural colorant has a lipophilic nature, its application in high water content foods is limited. Also, its bioavailability will reduce. Thus, the application of emulsion-based systems is suitable to overcome these challenges (Davidov-Pardo, Gumus, & McClements, 2016).

The emulsion-based delivery system is suitable because the lipid phase finally breaks down in the gastrointestinal system and forms the colloidal structures that then solubilize and transfer the bioactive component and increase its bioavailability (Davidov-Pardo *et al.*, 2016). One of the most important effects of emulsions is the big surface of the lipid phase that can help more digestion in the human intestinal area (Frede *et al.*, 2014).

The diversity of emulsion-based delivery systems is high and different types of them are used for encapsulating the bioactive components including association colloids, simple solutions, emulsions, biopolymer matrices, powders, and so on. However, delivery systems, especially for bioactive lipid components, should have different specifications (McClements *et al.*, 2007):

1- The ability to encapsulate the bioactive component into a physical form to incorporating in foods or beverages.
2- The compatibility with food or beverage that is incorporated into (no effect on color, flavor, texture, or shelf-life food product).
3- Protecting the bioactive component from degradation during preparation, transport, storage, and application in food products so that the active state of the bioactive component remains a state.
4- The capability to release the bioactive component at a certain rate and site or response to conditions like pH, ionic strength, and temperature.
5- Preparing from food-grade materials by simple, cost-effective, and practical method for use in food industries, biodegradability, regulatory status, biocompatibility, ease of use, and so on.

This method can be effective for natural colorants that have also the neutraceutical aspects behind the coloring role. But it should be noted that for industrial applications, the physical and chemical stability at different food formulation or storage conditions of this system should be undertaken.

Weigel *et al*. (2018) have used an oil-in-water emulsion to stabilize the lutein. Lutein is an oxygenated carotenoid with a yellow to red color that is mainly isolated from *marigold* flowers. Lutein is a hygroscope molecule with low water solubility and low oxidative stability. Weigel *et al.* (2018) made a *quillaja* saponin emulsion that contained 2.5% of lutein suspension (20% lutein in corn oil) dispersed in corn oil (which made up 5% of the total emulsion), the final amount of lutein presents in the emulsions was 250 mg/L. Then they have studied the impact of emulsifier type (*quillaja* saponin, Tween 80, whey protein, and casein) and antioxidant type (EDTA, ascorbic acid, catechin, α-tocopherol, and ascorbic acid palmitate) on the physical and chemical stability of lutein-fortified emulsions. They found during storage at 45 °C for 10 days, the emulsions prepared by *quillaja* saponin (as an emulsifier) and ascorbic acid (as an antioxidant) showed suitable color stabilization (Weigel *et al*., 2018). Davidov-Pardo *et al.* (2016) studied the effect of temperature (5-70 °C) and pH (2-8) on lutein-enriched emulsions during 7 and 14 days of storage. They found temperature increases the chemical degradation of lutein emulsion. And the pH has a great effect on the physical stability of emulsion (Davidov-Pardo *et al*., 2016).

Frede *et al.* (2014) examined the effect of six different emulsifier compositions on lutein bioavailability. They studied the β-lactoglobulin, β-lactoglobulin/lecithin, Biozate 1 (the whey protein hydrolysate), Biozate 1/lecithin, Tween 20, and Tween 20/lecithin on stability, cytotoxicity, and lutein uptake by HT29 cells (Frede *et al*., 2014). They found whey proteins (alone or in combination with lecithin) had the most effective results. Also, the type of emulsifier has an important role in lutein uptake and the combination of Biozate 1 and lecithin showed the highest uptake by HT29 cells (Frede *et al*., 2014).

Encapsulating is the most common method for stabilizing the wide range of natural colorants. The encapsulating of bioactive components with a polymer can protect them from oxygen, light, enzyme degradation, water, and other conditions (Ravichandran *et al*., 2014).

Encapsulation almost ensures the delivery of a determined amount of bioactive components to the human body. Also, it can promote easier handling, prevents lumping, improves flowability (Ravichandran *et al*., 2014). Different encapsulating materials like polysaccharides (starch, maltodextrins corn syrups, and Arabic gum), lipids, proteins (gelatin, casein, soy, and wheat protein) can be applied for encapsulating process (Ravichandran *et al*., 2014).

The encapsulating is done in different ways: (1) spray drying and spray chilling, (2) fluidized bed coating, (3) extrusion, (4) emulsification and liposomes, (5) spinning disk, and (6) coacervation (Kaimainen, 2014). Spray drying is used mostly in natural colorant encapsulation because this method is a low-cost and efficient method for natural colorant encapsulation (Kaimainen, 2014). Encapsulating the natural colorants is almost done during the drying step of natural colorant production (Weber *et al*., 2017). This leads to decreasing the exposure of natural colorant to oxygen, light, and some degrading factors and can increase the shelf life of natural colors. When a natural colorant is encapsulated, the handling, solubility, stability, and flow properties will improve, and the addition of nutrient to dry mixtures will reduce the dust arising from ingredients (Ravichandran *et al*., 2014). Ravichandran *et al.* (2014) studied the effect of different encapsulating agents like maltodextrin, guar gum, Arabic gum, pectin, and xanthan gum on betalain stability. They used spray drying and freeze-drying for comparison to the effect of drying methods in encapsulation. They found when the freeze-drying method and maltodextrin with xanthan and guar gum (as coating agent) are used, the protection for betalain will be high (Ravichandran *et al*., 2014).

Hernández-Martínez *et al.* (2017) encapsulated betalain red dye (extracted from *Beta vulgaris* L. (beet)) with tetraethyl orthosilicate (TEOS) and compared it with a commercial dye FD&C Red 40. They found the encapsulated betalain red dye is more resistant to UV light and less resistant against pH and temperature changes (Hernández-Martínez *et al*., 2017).

Astete *et al.* (2009) studied the effect of entrapment of β-carotene (lipophilic natural pigment) with a water-soluble mixture of Ca^{2+} cross-linked alginic acid. They found the calcium can increase the nanoparticle density and improve the colorant stability against oxidation (Astete *et al*., 2009).

Food formulations and additives

The most important factors affecting natural colorants stability in food formulation or storage are temperature, pH, water activity, oxygen, light, chelating agents, the presence of other compounds, pigment concentration, storage, and processing conditions (Natalia Martins *et al*., 2017), Brix (solid content) (Kırca, Özkan, & Cemeroğlu, 2007). Kırca *et al*. (2007) studied the effect of solid contents or Brix (11, 30, 45, and 64 °Brix), pH (4.3 and 6), and heating treatment (70-90 °C) on black carrot anthocyanin stability during storage at 4-37 °C (Kırca *et al*., 2007).

Some additives like citric acid can increase the stability of natural colorants (Pan-utai *et al*., 2017). Pan-utai *et al*. (2017) studied the effect of citric acid addition to C-phycocyanin (C-PC) (a blue pigment often found in cyanobacteria) during incubation at 80 °C for 1 h. They found citric acid can increase the stability of C-phycocyanin in high thermal processing applications and extraction (Pan-utai *et al*., 2017). Also, Martelli *et al*. (2014) investigated the effect of the addition of natural protein crosslinker methylglyoxal, honey, and a high concentration of sugar on C-phycocyanin. They found methylglyoxal does not a significant effect on color stabilization. Honey and a high concentration of sugar can play a preservative role in color stabilization. They believed that the preservative effect of a high concentration of sugar on the blue color of C-phycocyanin is dependent on the final concentration of sugar in solution rather than the type of sugar (Martelli *et al*., 2014). Sugars and their degraded products at low concentrations may decrease the stability of natural colorants like anthocyanins (Bechtold & Mussak, 2009). Fructose because of its high solubility, had the highest preservative effect, especially in saturated concentration. They also found the blue color diminishes a little after 2 months of storage when the fructose is used for the stabilization of color during sterilization at low (80 °C) and high (100 °C) temperatures (Martelli *et al*., 2014).

Betalains are more stable than anthocyanins and their stability will increase with an increase in color concentration, lowering the pH about 4-7, glycosylation, and acylation in high levels, reducing water activity, oxygen content, and temperature, and in presence of chelating agents, antioxidants and cyclodextrins (Rodriguez-Sanchez *et al*., 2017).

Rodriguez-Sanchez *et al*. (2017) investigated the effect of pH, temperature, gum formulation, and beverage formulation on betaxanthins stability and antioxidant properties. They have found the pH 6.6 and the presence of ascorbic acid as an additive showed the maximum stability of betaxanthins. They also found the betaxanthin application in gummies is more stable the color than in beverage formulations and this stabilization is more when the product is stored in darkness and low temperature (Rodriguez-Sanchez *et al*., 2017).

Calvo & Salvador (2000) have examined the stability of 4 natural colorants of annatto (orange), chlorophyllins (green), cochineal (red), and curcumin (yellow) during gel making. They made their samples with gelatin, a mixture of xanthan and locust bean gum, sugar, and natural colorant. Then they measure the color (with a Hunter Labscan II colorimeter) and sensory parameters (a team of 10 judges). They have found that the cochineal and curcumin natural colorants can replace the synthetic ones in jellies (Calvo & Salvador, 2000).

Duangmal *et al*. (2008) studied the color of freeze-dried roselle extract as a natural color in a beverage model system. They extracted the colorant by acidified water-95% ethanol (1:1) and then freeze-dried the extract. They used maltodextrin and trehalose as a stabilizer (a and 3 g/100 g extract). After freeze-drying, the amount of 0.1 g/100 model drink was used and then color stability was studied during 12 weeks storage at 30 °C (Duangmal *et al*., 2008). They found the lightness and hue changes were low during storage and maltodextrin had a higher stabilizing effect than trehalose. They also compared the roselle extract with SAN RED RC(R) and carmoisine in the model drink and found the hue of all three samples was the same but the color of the drink with roselle extract was not stable for more than 56 days (Duangmal *et al*., 2008).

(a) (b) (c)

Fig. 8. Colored yogurt (a), pieces of bread (b), and cake (c)

Table 2. Total phenolic contents (mg GAE/L) during 3 weeks

	Amount of extract	Week 1	Week 2	Week 3
Cake samples	0.5	612±5.07^{c}	599±3.09^{b}	560±8.03^{c}
	1.5	834±4.02^{b}	806±7.04^{a}	783±5.03^{b}
	2	917±6.01^{a}	894±4.08^{a}	880±4.05^{a}
Yogurt samples	0.5	612±0.01^{c}	599±0.01^{c}	560±0.01^{c}
	1.5	834±0.04^{b}	806±0.03^{b}	783±0.01^{b}
	2	917±0.04^{a}	894±0.04^{a}	880±0.04^{a}

Numbers with different letters across each column for each colorant show a significant difference at (P<0.05)

Table 3. The peroxide value of lycopene colored cakes during 3 weeks

	Amount of extract	Week 1	Week 2	Week 3
Control	0	2.41±0.80^{a}	3.33±0.60^{a}	3.61±0.10^{a}
Lycopene oleoresin	0.5	0.61±0.40^{b}	0.77±0.30^{b}	2.54±0.30^{c}
	1.5	0.4±0.02^{c}	0.77±0.30^{b}	3.2±0.60^{b}
	2	0.57±0.70^{b}	0.89±0.10^{b}	2.05±0.30^{d}

Numbers with different letters across each column for each colorant show a significant difference at (P<0.05)

In fruit juice industries, some enzymes like pectinase are used for the purification of juice. Sometimes pectinases are used to increase the color extraction efficiency. But enzymes (glycosidases, peroxidases, and phenolases) can degrade anthocyanins and other pigments via hydrolyzing glycoside substitutes and make them unstable (Bechtold & Mussak, 2009). Ranjbar Nedamani (2020) used the natural colorant of lycopene oleoresin, chlorophyll oleoresin, and *Berberis Voulgaris* extract in bread, cake, and yogurt (Fig. 8). They measured the antioxidant activity of lycopene in cakes, total phenol content of *Berberis Voulgaris* extract in bread and yogurt. They used 0, 0.5, 1.5, and 2% levels of colorants. They found in yogurt, the total phenol contents of samples were reduced after 3 weeks. Also, the total phenols of cake samples were not significantly changed after 3 weeks (Table 2). Also, lycopene oleoresin showed significant antioxidant activity in cakes. Also, the activity was high at weeks 1 and 2, but it reduces at week 3 (Table 3) (Ranjbar Nedamani, 2020).

Extraction with green solvents

Dai, Verpoorte, & Choi (2014) proposed natural ionic solvents and natural deep eutectic solvents (NADES) as health solvents in food, pharmaceutical, and cosmetic applications. These solvents have advantages over synthetic ionic solvents. They have a unique power to dissolve non-water-soluble components. They are low cost, biodegradable, non-toxic, sustainable, and simple preparative, negligible volatility, liquid form even at below temperatures than 0, adjustable viscosity, wide polarity range, and high solubility for a wide range of components (Dai *et al.*, 2014). NADES can be stabilizing media for solutes. They are liquid supermolecules made of natural primary metabolites that intermolecular interact (hydrogen bonds) together (Dai *et *al*., 2014). NADES can be found in cell membranes, and involved in biosynthesis, solubilization, and storage of poorly water-soluble/unstable components in cells (Dai *et al*., 2014). This leads to arising ideas that explain the stability of natural colorants in plants (Dai *et al*., 2014). Dai *et al*. (2014) showed safflower pigments are more stable in sugar-based NADES than water or 40% ethanol solution due to hydrogen bonding between NADES molecules and solute. This ability also can be adjusted by reducing the water activity and viscosity increasing (Dai *et al*., 2014).

Storage conditions

Different mechanisms can help the stabilization of the natural colorants after the extraction. Conditions such as controlled atmosphere, drying, and etc. can increase the color (Cortez *et al.*, 2017; Topuz, Feng, & Kushad, 2009). Howard *et al*. (2014) determined the retention of color and anthocyanins of strawberry puree prepared under nitrogen, CO_2, and air. They pasteurized the samples and stored them at 25 °C. The color evaluation was done at to week intervals for 8 weeks. They found the samples prepared at nitrogen or CO_2 conditions have more anthocyanins and color stability (Howard *et al*., 2014).

Conclusions

Human health is the most important aspect of food production in industries. With the growing knowledge of consumers, the application of synthetic ingredients in food products decreases. The substitution of flavors, colors, odors, and so on with natural ones, is big progress to find new formulations in food products and consumers. Natural colorants behind the coloring ability can play roles in human health, and some of them like carotenoids can have an antioxidative effect during the storage of foods. Also, some like lycopene showed antibacterial effects against some important pathogenic microorganisms. These aspects make natural colorant a good ingredient in food formulations. But their stability according to different food production and formulation factors should be considered. Also, it should be mentioned that the stabilization methods should be chosen to have the least side effects occur.

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افزایش پایداری رنگدانه‌های طبیعی مواد غذایی- مقالۀ مروری

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چکیده

آگاهی عمومی دربارۀ اهمیت کاربرد اجزای طبیعی در محصولات غذایی، حوزۀ جدیدی در استخراج، پایداری، نگهداری و کاربرد رنگ‌های طبیعی ایجاد کرده است. پایداری رنگ‌های طبیعی نقش کلیدی در کاربرد آنها در صنایع غذایی دارد. رنگ‌ها در خارج از منابع طبیعی خود به شکل بارزی حساس بوده و کاهش رنگ برخی از آنها بلافاصله بعد از استخراج آغاز می‌شود. در این مقاله، روش‌های کاربردی رایج برای پایداری رنگ‌های طبیعی در صنایع غذایی مرور شد. هدف این مقاله، مرور نتایج علمی منتشرشده دربارۀ روش‌های پایداری رنگ‌های طبیعی مختلف می‌باشد. منابع اطلاعاتی گوگل اسکولار، پاب‌مد، وب آو ساینس موردبررسی قرار گرفتند. درنهایت حدود 120 مقالۀ علمی انتخاب‌شده و 73 مقاله مرتبط مورد استفاده قرار گرفتند. این مرور از مقالاتی که در سال 2020 منتشرشده بودند، آغاز شد و تا مقالات سال 2000 ادامه یافت. دربارۀ روش‌های پایدارکردن رنگ‌های عمدۀ طبیعی مانند آنتوسیانی‌ها، کاروتنوئیدها، کلروفیل و بتالایین گزارش‌های زیادی ارائه شده است. این روش‌ها را می‌توان قبل، در طول و بعد از فرمولاسیون و تولید محصولات غذایی به‌کار برد. به‌دلیل نقش‌های متعددی که رنگ‌های طبیعی در سلامت انسان و همچنین پایداری اکسیداتیو محصولات غذایی ایفاء می‌کنند، پایداری و کاربرد رنگ‌های طبیعی در محصولات غذایی می‌تواند یک انتخاب مناسب در صنایع غذایی باشد.

واژه‌های کلیدی: انکپسولاسیون، پایدارکردن، تشکیل کمپلکس، رنگ طبیعی، فعالیت کوپیگمنتاسیون