Alternatives to Those Artificial FD&C Food Colorants

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Abstract

Replacement of artificial food dyes with natural colorants is a current marketing trend, notwithstanding the fact that neither the United States nor the European Union (EU) has defined natural with respect to food colors. Consumer groups have concerns over the safety of synthetic colorants, and in addition, many of the naturally derived colorants provide health benefits. Food scientists frequently have the assignment of replacing artificial colorants with natural alternatives. This can be challenging, as naturally derived colorants are usually less stable, and all desired hues might, in fact, not be obtainable. In this review, the chemical and physical properties, limitations, and more suitable applications for those colorants that are legally available as substitutes for the synthetic colorants are summarized. Issues and challenges for certain foods are discussed, and in addition, colorants that may be available in the future are briefly described.

INTRODUCTION

GRAS: generally recognized as safe

FAO: Food and Agricultural Organization

WHO: World Health Organization

JECFA: Joint FAO/WHO Expert Committee on Food Additives

FD&C: food, drug, and cosmetic

GHA: global hyperactivity aggregate

ADI: acceptable daily intake

EU: European Union

FDA: Food and Drug Administration

Coloring substances have been used to enhance the appeal of foods since the beginning of recorded history (Marmion 1991, Arit 2011). The indiscriminate use of a variety of dyes that were subsequently found to be toxic or poisonous influenced the passage of legislation, such as the Food and Drug Act of 1906. Other major legislation in the United States concerning food colorants includes the Food Drug and Cosmetic Act of 1938 and the Color Additives Amendment of 1960 (Francis 1999). Current regulations of color additives are based on two principles: (*a*) the additive must not be harmful, and (*b*) it must not mislead consumers. All color additives must be on a positive list of approved applications and used with specified levels. Terms such as GRAS (generally recognized as safe) and nature-identical do not apply to color additives. Regulations on colorant use can vary markedly from country to country. Many countries have adopted the approved applications and usage levels posted by the Codex Alimentarius (Joint FAO/WHO Codex Alimentarius Commission 2008) and color additive specifications published by the Joint FAO (Food and Agricultural Organization)/WHO (World Health Organization) Expert Committee on Food Additives (JECFA) (2011). Other countries regulate independently the use of color additives. Examples include the United States (Code of Federal Regulations 2011), Korea (Korea Food and Drug Administration 2004), and Japan (Japan Food Chemical Research Foundation 2006).

Evaluation of the safety of color additives receives particular scrutiny, and the subject has been summarized in several reviews (Marmion 1991, EFSA 2008, Food Advisory Committee 2011). The possible allergenicity of food, drug, and cosmetic (FD&C) Yellow Number 5 (Tartrazine) caused consumer concern in the 1980s, which had an impact on food labeling (Food Advisory Committee 2011) and stimulated some processors to convert to natural colorants. The possible link between hyperactivity in children and the consumption of artificial food colorants has been revisited in the Southampton study (McCann et al. 2007). The effect of consuming a blend of synthetic food colors on the behavior of groups of three-year-old and eight- to nine-year-old children was investigated, and the authors concluded that the global hyperactivity aggregate (GHA) score increased for some groups of children consuming one mix of colorants compared with those consuming a placebo. The methods and conclusions from the Southampton study were reviewed by a panel convened by the European Food Safety Authority (Scientific Opinion 2008), which concluded, "The findings of the study cannot be used as a basis for altering the ADI (acceptable daily intake) of the respective food colours or sodium benzoate." Despite the panel's report, foods and beverages containing any of the Southampton dyes sold in the European Union (EU) after July 2010 must contain a warning label: [name or E number of color(s)] may have an adverse effect on activity and attention in children. The Food Advisory Committee of the Food and Drug Administration (FDA) recently conducted a hearing on certified color additives and their possible association with hyperactivity in children (Food Advisory Committee 2011). Their review of published studies led them to conclude "a causal relationship between exposure to color additives and hyperactivity in children in the general population has not been established." Members of the FDA advisory panel noted, however, that there may be a relationship between color additives and a small group of children, but existing scientific data established neither the relationship nor the identity of the children, and as a result the FDA should take no action. The hearing and its conclusions have had a major impact in the marketplace with some consumer groups and corporations insisting that artificial food dyes be replaced with natural colorants. Neither the EU nor the FDA has defined the term natural in relation to food colors. The Natural Food Colours Association (2011) has suggested extending the EU definition of natural flavor to food colors. It is unlikely, however, that a global definition of natural color will be adopted in the near future because of the lack of consensus on how consumers interpret the term.

A frequent assignment for food scientists is to replace certified colorants with more label-friendly natural colorants in food formulations. This task can be quite challenging. Naturally derived colorants are usually less stable to heat, light, and oxygen. They may interact with other ingredients, resulting in the development of unwanted colors and flavors. Naturally derived colors are usually less vivid. An appealing feature of natural colorants is that many may provide health benefits. Alternative color systems do not exist for all hues, e.g., FD&C Blue Number 1. Most of these challenges can be met with careful product design and testing. Also, it may be necessary to modify packaging and/or shorten product shelf life. An objective of this review is to summarize the alternatives available for replacing artificial colors with naturally derived colorants and to describe their limitations and suitability for different food applications.

THE RED HUES

Anthocyanins

Anthocyanin pigments are not a single chemical entity; rather, they are a class of compounds, with more than 600 anthocyanins having been identified in nature (Anderson & Jordheim 2008). A generalized structure for anthocyanins is shown in **Figure 1**. Structural variation comes from various patterns of substitution in the B ring with OH and OCH_3 groups, presence of different sugar substituents at the 3 and 5 positions, and the possibility of acylation of sugar substituents with cinnamic and aliphatic acids. These structural variations have an impact on both color and stability. An increased number of OH and OCH_3 groups will shift the hue from red to bluish-red to purple. The color of anthocyanin-pigmented fruits and vegetables can vary from the intense red of a strawberry to the glossy purple of an eggplant and the dark blue of a blueberry. Extracts and juices

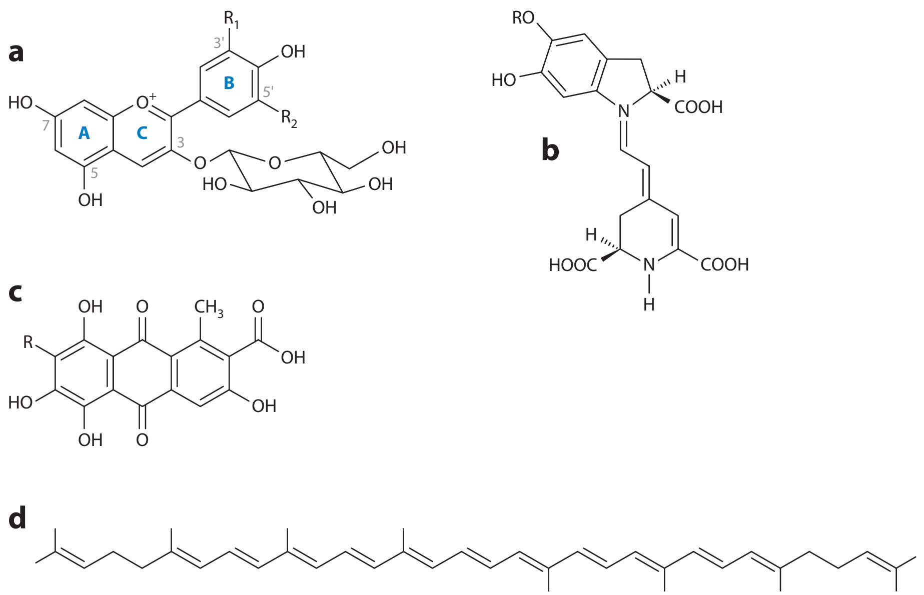

Figure 1

The red hues. Generalized structure for (*a*) anthocyanins, [R_1 and R_2 = H, OH, or OMe; glycosidic substitution on 3, 5, or 7; possible acylation on sugar(s)], (*b*) betacyanins (R = glucose = betanin; R = H = betanidin), (*c*) carminic acid, and (*d*) lycopene.

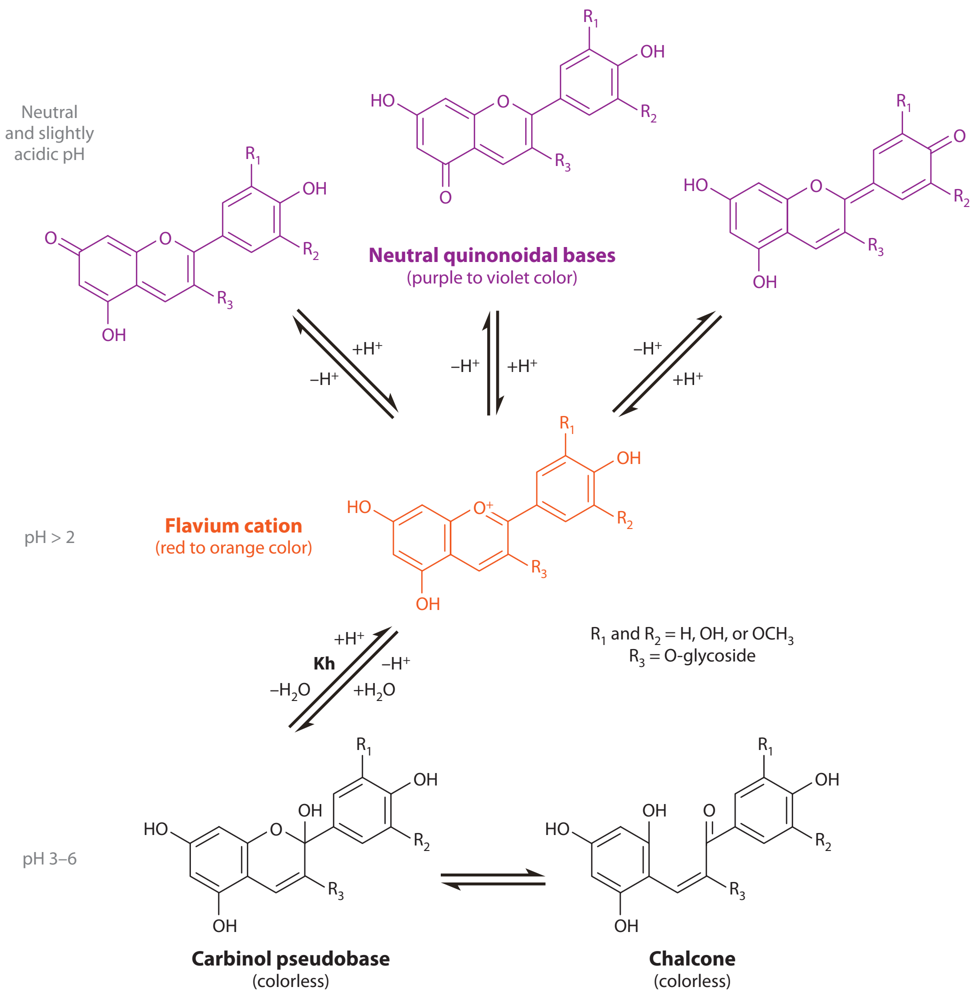

Figure 2

Scheme of the pH-dependent structural interconversion between dominant forms of monoglycosylated anthocyanins in aqueous phase. (Houbiers et al. 1998) Adapted from He & Giusti 2010.

of these plant materials do not necessarily exhibit the same color as the starting products because both chemical and physical factors can markedly affect color and appearance. Anthocyanins are soluble in water and alcohol. They are reactive compounds, being susceptible to degradation by both heat and light exposure. Anthocyanins condense with other phenolic compounds to form polymeric pigments. Thus, the color of anthocyanin-based colorants can be substantially due to polymeric pigments, as is the case for most grape-skin extract samples. Increased glycosidic substitution, and acylation of sugar groups with cinnamic acids, in particular, improves the stability of anthocyanins.

Anthocyanins reversibly undergo a number of structural changes with change in pH (**Figure 2**), which has a major impact on color because the color of these forms varies from red to colorless to purple to blue. The different forms also vary in their stability; hence, pH also affects degradation rate. At a nonfood pH of 1, effectively 100% of anthocyanins are in the colored flavylium form, usually expressed as an intense red color. As pH is increased to a typical beverage pH of 3.5, the intensity decreases, but an attractive red hue persists. With further increase in pH, the color fades and a shift to a bluish hue occurs. Acylation affects the pH at which these transformations occur so that acylated anthocyanins, such as those found in black carrot, red radish, purple sweet potato, and red cabbage, can provide acceptable color at a pH of 4.5 or even 5. The acylated anthocyanins are also more stable to light and heat. In addition to pH, the water activity of a food system affects degradation rate; the lower the water activity, the greater the stability (Amr & Al-Tamimi 2007, Erlandson & Wrolstad 1972, Garzon & Wrolstad 2001). For a more definitive treatment of anthocyanin chemistry, see the following references (Schwartz et al. 2008, Anderson & Jordheim 2006, Wrolstad 2000, Mazza & Miniati 1993).

Several anthocyanin-based colorants approved for food use are listed in **Table 1**. In the United States, these are classified under the category of color additives that are exempt from certification (Code of Federal Regulations 2011), whereas in the EU anthocyanin-derived colorants are categorized under classification number E 163. Anthocyanin colorants are also recognized as being appropriate for food use in Asian, Central American, and South American countries; however, there are differences with respect to what sources and degree of purity are permitted (Culver & Wrolstad 2008). Grape-skin extract has been used as a colorant for more than one hundred years, its first application being to enhance wine color (Francis 1999). Because anthocyanins are concentrated in the peel, extraction of skins as opposed to the whole fruit allows for pigment concentration. Fermentation to remove any sugars further concentrates the pigment. The anthocyanins undergo polymerization reactions during extraction and fermentation so that the resulting product is quite high in polymeric color. Grape-color extract is obtained as a byproduct from the juice processing of Concord grapes, *Vitis labruscana*. The original petition for its use as a colorant in the United States limited its application to nonbeverage food use, and that limitation is still in place (Code of Federal Regulations 2011). The remaining anthocyanin colorants are approved in the United States under the categories of fruit and vegetable juices. Aqueous extraction of dried fruits and vegetables is also permitted in this category. These colorants are widely used in nonalcoholic and alcoholic beverages. They are generally unsuitable for milk-based beverages because of the change to bluish hues at milk's pH of 6.7. They are not well suited for extruded products because of their tendency to undergo heat degradation. The preponderance of acylated pigments in vegetable-based anthocyanin colorants (black carrot, radish, purple sweet potato, red cabbage) imparts greater heat and light stability to the products. Hence, they are less susceptible to heat degradation in extrusion processing. Anthocyanins' stability at low water activity makes them good candidates for dry-mix beverages and some snack foods. There is considerable variation as to the hue and stability of the different anthocyanin colorants. Therefore, a range or even a combination of anthocyanin colorants should be evaluated for specific product applications. Another attractive property of anthocyanins is their possible health benefits. There is substantial evidence that dietary consumption of anthocyanins may provide anti-inflammatory and anticarcinogenic activities, reduce the incidence of cardiovascular disease, and help in obesity control and diabetes alleviation (He & Giusti 2010).

Betacyanins

This class of pigments is not widely distributed in nature, and the source for their use as a food colorant is presently limited to red beets (*Beta vulgaris* sp.). Both beet juice concentrate and beet powder are approved colorants (Code of Federal Regulations 2011). A generalized structure for betacyanin pigments is shown in **Figure 1**. The betacyanins are similar to anthocyanins in that they are water-soluble, red-colored phenolic glycosides. Their biosynthetic pathways, however, are

Table 1 Approved natural colorants available for food use

Colorant	Hue	21 CFR[a] Reference Number	E No.[b]	Suitable pH range	Stability		Restrictions, limitations
					Heat	Light	
Anthocyanin-based colorants							
Grape-skin extract	Red to purple	73.17	E 163	<3.5	Fair-good	Fair-good	Limited to beverages in the United States
Grape color extract	Red to purple	73.169	E 163	<3.5	Fair-good	Fair-good	For nonbeverage use in the United States
Fruit juices							
Red grape	Red to purple	73.25	E 163	<3.5	Fair-good	Fair-good	
Blackcurrant	Red to purple	73.25	E 163	<3.5	Fair-good	Fair-good	
Elderberry	Red to purple	73.25	E 163	<3.5	Fair-good	Fair-good	
Chokeberry (*Aronia*)	Red to purple	73.25	E 163	<3.5	Fair-good	Fair-good	
Vegetable juices							
Black carrot	Red to purple	73.26	E 163	<4.5	Good-excellent	Good-excellent	
Red cabbage	Red to purple	73.26	E 163	<4.5	Good-excellent	Good-excellent	
Red radish	Red-orange to purple	73.26	E 163	<4.5	Good-excellent	Good-excellent	
Purple sweet potato	Red to purple	73.26	E 163	<4.5	Good-excellent	Good-excellent	
Purple corn	Red-orange to purple	73.26	E 163	<4.5	Good	Good	
Betacyanin-based colorants							
Beet powder	Red to red-purple	73.4	E 162	2.5–7	Poor	Poor	
Beet juice	Red to red-purple	73.26	E 162	2.5–7	Poor	Poor	
Carminic acid–based colorants							
Cochineal	Orange to red	73.1	E 120	3.5–8.0	Excellent	Excellent	Non-kosher
Carmine	Orange to red	73.1	E 120	3.5–8.0	Excellent	Excellent	Non-kosher

Carotenoid-based colorants							
Annatto	Yellow to orange	73.3	E 160b	3.0–8.0	Good	Fair-good	
β-apo-8′-carotenal	Orange	73.9	E 160e	3.0–8.0	Good	Fair-good	<15 mg lb^{-1} of solid/semisolid food or 15 mg per pint of liquid
Astaxanthin	Orange-red	73.35		3.0–8.0	Good	Fair-good	<80 mg kg^{-1} fish feed
Canthaxanthin	Orange-red	73.75	E 161a	3.0–8.0	Good	Fair-good	<30 mg lb^{-1} of solid/semisolid food or pint of liquid
β-carotene	Yellow to orange	73.95	E 160a	3.0–8.0	Good	Fair-good	
Carrot oil	Yellow to orange	73.3		3.0–8.0	Good	Fair-good	
Corn endosperm oil	Yellow	73.315			Good	Fair-good	Limited to use in chicken feed
Dried algal meal	Yellow	73.275			Good	Fair-good	Limited to use in chicken feed
Haematococcus algae meal	Orange to red	73.35			Good	Fair-good	Limited to <80 mg kg^{-1} salmonid fish feed
Paprika oleoresin	Yellow to orange-red	73.345	E 160c	3.0–8.0	Good	Fair-good	
Phaffia yeast	Yellow to orange-red	73.355			Good	Fair-good	Limited to <80mg kg^{-1} salmonid fish feed
Saffron	Yellow	73.5		3.0–8.0	Good	Good	
Curcumin-based colorants							
Turmeric; turmeric oleoresin	Bright yellow	73.6	E 100	3.0–7.0	Very good	Poor	
Chlorophyll-based colorants							
Na-Cu chlorophyllin	Green	73.125	E 141	3.0–7.0	Good	Good	<0.2% dry beverage mix
Melanoidin-based colorants							
Caramel	Brown	73.85	E 150a-d	3.0–7.5	Very good	Very good	
Titanium-based colorants							
Titanium dioxide	White	73.575	E 171		Excellent	Excellent	
Pearlescent pigments	Luster	73.35			Excellent	Excellent	

[a]Code of Federal Regulations (CFR).
[b]European Union system for designating approved food colorants (E No.).

quite dissimilar. Betacyanins have higher solubility in water than alcohol, whereas for anthocyanins the reverse is true. In contrast to the anthocyanins, the betacyanins are red colored throughout the pH range of 1–7 because they do not undergo the structural transformations analogous to the anthocyanins. The betacyanin pigments are very labile to light and heat degradation. This tends to limit their usage to foods with a short shelf life, and foods, such as frozen desserts, that are stored at low temperatures in opaque packaging. Their red color at a pH of 6–7 makes them good candidates for coloring dairy products, such as yogurts and ice creams. Beets have a distinctive earthy flavor, which is greatly reduced during concentration and dehydration. Preparations may contain some residual flavors that may be difficult to mask in delicately flavored products, such as certain confections. Betacyanins are effective free radical scavengers and have high antioxidant capacities (Stintzing & Carle 2004). The impact of these properties on human health from dietary consumption has not been extensively studied.

Cochineal and Carmine

Cochineal is the aqueous ethanol extract of the female cochineal insect *Dactylopius coccus* Costa, which lives on cacti from the genus *Opuntia*. The principal pigment is carminic acid (**Figure 1**), which is an anthraquinone. Carmine is a more purified colorant prepared from cochineal by boiling cochineal in ammonia or sodium carbonate. After filtration, carminic acid is treated with alum to form the calcium or calcium-aluminum lake of carminic acid, which is known as carmine. An aqueous solution of carminic acid at pH 4 is yellow to orange colored; however, when it complexes with aluminum and calcium, colors from magenta red to purple are produced (Schul 2000, Francis 1999). The textile industry uses other metals, such as tin, chromium, iron, etc., to obtain various colors for dyeing fabrics. For food use, the substrates are limited to aluminum and calcium. Carmine is insoluble in alcohol and has very limited solubility in water. It is extremely stable to light and heat, resistant to oxidation, and not affected by SO_2. Its affinity for protein is a desirable property for coloring surimi and comminuted meat products. In addition to its relative high cost, a major limitation for its use in foods is its not being kosher because of its insect source. Additional food applications are its use in beverages and fruit preparations, such as jams and preserves. Reports of possible allergenicity have been an issue with cochineal's usage; it is believed that the potential allergens are other protein contaminants present in the extract that can be removed (Delgado-Vargas & Paredes-López 2003). A wide range of cochineal and carmine preparations are available commercially for food applications, which vary as to carminic acid content, purity, calcium and aluminum content, and the presence of other solvent or carrier ingredients. Cochineal has a fascinating history regarding the discovery of its use in art, cosmetics, textiles, and foods in Mexico and South America by European explorers, and its subsequent adoption in Europe (Schul 2000).

Tomato Lycopene Extract

The acyclic carotenoid pigment lycopene (**Figure 1**) is the major pigment responsible for the bright red coloration of tomatoes. The hydrocarbon is insoluble in water and soluble in nonpolar solvents. Physical factors, i.e., the manner in which light is reflected from the pigment matrix in the tomato peel, contribute significantly to the appearance of the tomato fruit. A solution of lycopene in a solvent such as acetone will be orange colored and have a very different visual appearance from the tomato fruit. Tomato lycopene extract and tomato lycopene concentrate are approved for use as colorants exempt from certification (Code of Federal Regulations 2011). Oleoresins, powders, and water-dispersible preparations that can impart colors from yellow to orange to red are commercially available. The color is stable through a broad pH range, and it is quite temperature stable in the absence of oxygen. It is susceptible to oxidation, and coatings are used to protect the pigment from oxidation and light degradation. It is approved for use in foods in the United States, except for those products having a standard of identity, such as tomato paste and tomato sauce. Although lycopene does not have vitamin A activity, it is a powerful quencher of singlet oxygen. There is epidemiological evidence that a high intake of lycopene is associated with reduced risk of prostate cancer (Basu et al. 2010, Giovannucci 2005).

THE YELLOW AND ORANGE HUES

Carotenoids

The pigment lycopene, discussed above, is one of more than 700 carotenoid pigments that have been identified in nature (Schwartz et al. 2008). Carotenoids are the most widespread natural pigments, being present in photosynthetic and nonphotosynthetic tissues of higher plants, algae, fungi, bacteria, birds, fish, insects, and invertebrates. Animals do not synthesize carotenoids; rather, plants in their diet are the sources of the carotenoids found in milk fat, egg yolk, crustaceans, and salmonids. **Figure 3** contains the structures of some selected carotenoids that have particular application to food colorant usage. They have a 40-carbon isoprenoid structure, and are soluble in nonpolar solvents and insoluble in water. Some such as β-carotene and lycopene are hydrocarbons, which are classified as carotenes. Some are acylic, e.g., lycopene, whereas others contain terminal ring structures, e.g., β-carotene. The carotenoids containing oxygen functional groups, i.e., alcohol, keto, and aldehyde, are classified as xanthophylls. All have an extensive system of conjugated single and double bonds, but they can differ as to the number of double bonds, as to whether they are in conjugation with carbonyl groups, and as to whether they contain a ring structure. These structural variations account for a range in the wavelength of maximum absorption in the visible spectrum approximating 440–489 nm, which results in hues from yellow to orange-red. Pigment concentration also influences the expressed color so that the same pigment may appear yellow to orange depending on its concentration. Carotenoids that have an unsubstituted β-ionone ring, such as β-carotene, possess vitamin A activity. The carotenoid pigments have varying degrees of antioxidant activity that may contribute significantly to human health. A downside to this desirable property is that they are readily oxidized, which can lead to bleaching and off-flavor formation. Hence, carotenoid colorant preparations often have protective coatings and/or antioxidant ingredients to protect the pigment from degradation.

Most often, β-carotene, β-apo-8′-carotenal, canthaxanthin, and astaxanthin are manufactured by chemical synthesis; they are classified in the United States as colorants that are exempt from certification (Code of Federal Regulations 2011). Natural sources of β-carotene from the algae *Dunaliella salina* or the fungus *Blakeslea trispora* are also available. Although the use of β-carotene is per good manufacturing practices, canthaxanthin and β-apo-8′-carotenal have restrictions on the quantity that can be used in foods (**Table 1**). Astaxanthin is limited to animal feed formulations and is widely used for farm-ranched salmon. Its usage in the United States requires a declaration of "color added."

Powdered and oil-based carotenoid preparations are available for use in lipid systems, such as margarines and oils. Water-dispersible formulations are commercially available, and some preparations have pigment particle size sufficiently small so that the appearance of an aqueous formulation approaches visual clarity.

Paprika oleoresin is prepared by solvent extraction of dried red sweet peppers, *Capsicum annuum* (Locey & Guzinski 2000). The extract contains a mixture of different carotenoids along with other lipid material. Both oil-soluble and water-dispersible preparations are available.

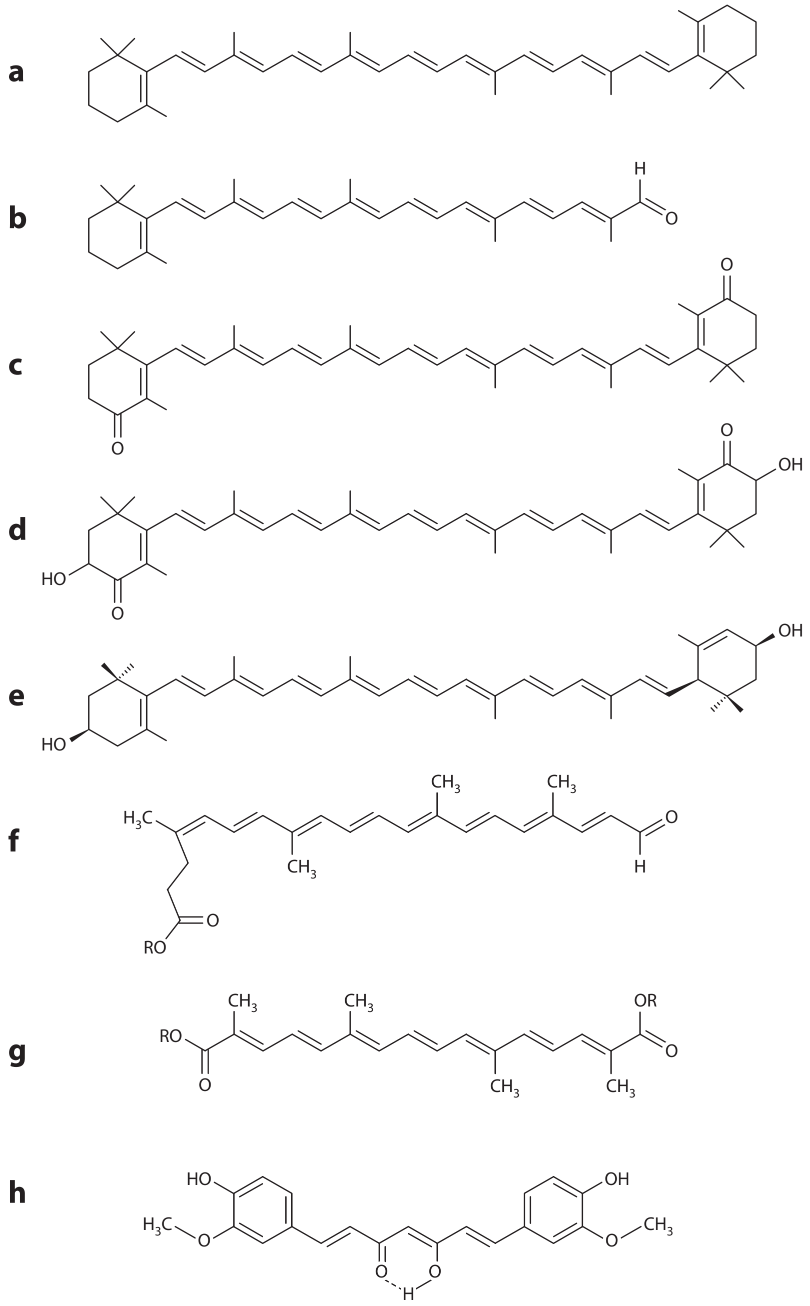

Figure 3

The yellow to orange hues. Structures of selected carotenoids. (*a*) β-carotene, (*b*) β-apo-8′-carotenal, (*c*) canthaxanthin, (*d*) astaxanthin, (*e*) lutein, (*f*) bixin (R = CH_3), norbixin (R = H), (*g*) crocin (R = gentiobiose), and (*h*) curcumin.

Formulations differ as to pigment content, and characteristic flavors may be present or removed through deodorization for different food applications. Carrot oil is another extract approved for use as a food colorant that is rich in β-carotene. Tagetes (Aztec marigold) meal and extract is restricted to usage in poultry feed to enhance the yellow color of chicken skin and eggs. It is a very rich source of lutein (**Figure 3**), which along with zeaxathin (present in paprika oleoresin) has a very important role in reducing the risk of age-related macular degeneration (Delcourt et al. 2006). The pigments in situ are mostly esterified to fatty acids. Marigold extracts with lutein concentrations as high as 20% are approved for use as dietary supplements in the United States, but not for use as a food colorant. In the EU, lutein preparations are an approved food additive, E161b. Additional carotenoid extracts (corn endosperm oil and dried algae meal) have approval for use in poultry feed and fish (salmonid) feed (*Haematoccus* algae meal, *Phaffia* yeast); refer to **Table 1**.

Annatto and Saffron

Annatto has widespread usage as a yellow to orange food colorant. It is particularly suitable for many dairy and bakery applications. Annatto's source is the seed of the tropical tree *Bixa orellana*, which grows in Central and South America. Centuries ago, European explorers discovered the aboriginal use of the seed for food, cosmetic, and religious purposes (Levy & Rivadeneira 2000). Annatto seeds and extracts have been used as a food colorant in Europe and North America for more than 100 years. Extracts are prepared by solvent extraction of the seeds. Bixin (**Figure 3**) is the major pigment in the annatto seed. It has the characteristic isoprenoid structure of the carotenoid pigments discussed above, but has a much shorter chain length, having 26 carbons as opposed to 40. The lipid-soluble compound has a carboxylic acid group at one end, and a methyl ester at the opposite end. Saponification of the methyl ester produces the water-soluble pigment norbixin (**Figure 3**). Thus, both water-soluble and oil-soluble annatto preparations are available. Bixin and norbixin contain a *cis* configuration as illustrated in **Figure 3**. Heat treatment can result in formation of the *trans* isomer, which is more lipid soluble and shows a color shift from red to more yellow tones (Levy & Rivadeneira 2000, Francis 1999). Commercial preparations are available in liquid and powdered forms, and they vary as to bixin, norbixin, and *trans*-norbixin contents, pH, and the presence of antioxidants, emulsifiers, and maltodextrin drying aids. There are preparations designed for use in acid foods, as low pH can lead to pigment isomerization and pinking. Many of these preparations contain synthetic emulsifiers that may be objectionable to some food companies; hence, it is critical that the identity of all diluents and emulsifiers in the acid proof formulation be understood.

The intense yellow color of the spice saffron is primarily due to the pigment crocin (**Figure 3**), where the C-20 dicarboxylic acid pigment crocetin is esterified with two molecules of the disaccharide gentiobiose. The sugar substituents render the pigment water soluble, and its use as a colorant has approval in the United States. Saffron consists of the dried stigmas of the flowers of the crocus bulb, *Crocus sativus*. Production of saffron is very labor intensive, and the extremely high cost of the spice severely limits its use as a food colorant. The saffron pigments are quite stable to light and heat. Crocin, along with other crocetin glycosides, is found in the fruit of *Gardenia jasminoides* and *Gardenia augusta* (Kato 2000). Gardenia yellow is a colorant produced by water or ethanol extraction of the fruits and is approved for food use in Japan and China, but not in the United States or Europe.

Turmeric and Turmeric Oleoresin

Curcumin (**Figure 3**) is the major yellow pigment in the spice turmeric. Two additional pigments that accompany curcumin are demethoxycurcumin, in which one methoxyl group is replaced with a proton, and bisdemethoxycurcumin, in which both methoxyl groups are replaced with protons. The pigments are lipid soluble and water insoluble. Rhizomes produced by the turmeric plant *Curcuma longa* are dried and ground to produce the spice that contributes to the distinctive color and flavor of mustards, pickles, and curry powder. Turmeric oleoresin is an approved colorant (Code of Federal Regulations 2011) that is prepared by solvent extraction of turmeric powder. Alternate processes are used with different solvents to produce oleoresins with varying pigment and flavor content (Buescher & Yang 2000, Pintea 2008b). Lipid-soluble and water-dispersible preparations, as well as dried powders, are commercially available with varying curcumin content. The expressed color is bright yellow or greenish-yellow. The pigment is unstable to light, and susceptible to oxidation. It is reasonably heat-stable and successfully used in extruded foods. Typical applications are margarines and oil/fat products, dairy and bakery products, dry mixes, and cereals. There has been extensive research on the anticarcinogenic properties of curcumin (Aggarwal et al. 2003). Curcumin has been described as a potent antioxidant and anti-inflammatory agent. There is evidence that curcumin can suppress tumor initiation, promotion, and metastasis (Aggarwal et al. 2003).

Riboflavin

The water-soluble B-complex vitamin riboflavin is responsible for the yellow to orange color of vitamin supplements. The synthetic vitamin is approved for use as a food colorant in the United States (Code of Federal Regulations 2011) and the EU. The EU also accepts usage of riboflavin-5′-phosphate as a food colorant, but the United States does not (Francis 1999). It is unstable to light and susceptible to oxidation. Its use as a colorant is largely for fortified cereals and dairy products.

THE GREEN AND BLUE HUES

Chlorophyll

Selection of a natural green food colorant is limited to derivatives of chlorophyll, the pigment of green plants, algae, and some bacteria that plays such an important role in photosynthesis (Hendry 1996a). There are two key structural features of the chlorophyll pigments (**Figure 4**) that relate to its solubility and color properties. The long chain isoprenoid alcohol phytol, which is esterified to a carboxylic acid functional group, gives the pigments lipid solubility. Saponification with strong alkali removes phytol, producing the water-soluble pigment chlorophyllin. The magnesium ion is easily displaced with two protons to form the olive-green pheophytin pigments (Schwartz et al. 2008). This reaction is irreversible; however, pheophytin will complex with copper and zinc ions to form complexes that have a more attractive green color and are more stable to light. Sodium copper chlorophyllin is an approved food colorant in the United States that is exempt from certification (Code of Federal Regulations 2011); however, at present its usage is restricted to dry mix citrus-based beverages at a level not exceeding 0.2%. The EU has approved use of copper chlorophyllin and copper chlorophyll complexes in a broad range of foodstuffs (Socaciu 2008b). Dried lucerne (alfalfa) is the primary source for production of the colorant. After solvent extraction, the crude extract is saponified to form water-soluble derivatives that are subsequently acidified in the presence of copper salts to form the stable green colorant (Hendry 1996a). Chlorophyllin is also used orally as an internal deodorant and topically in the treatment of slow-healing wounds (Higdon & Drake 2009). Clinical trials have shown that oral consumption of chlorophyllin is effective in

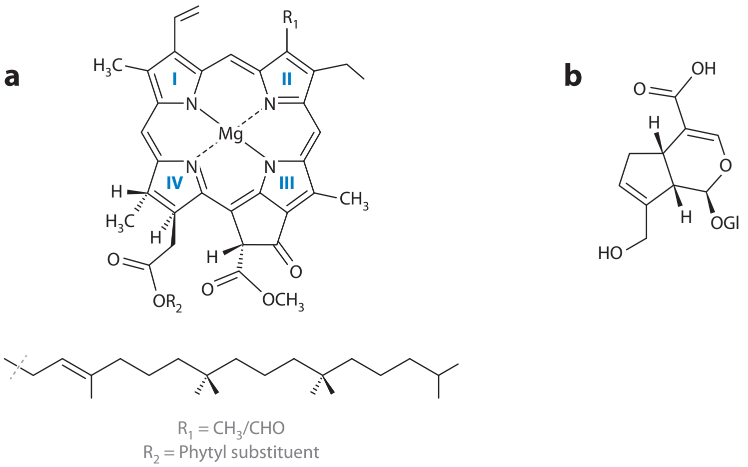

Figure 4

The green and blue hues. Structures of (*a*) chlorophyll a ($R_1 = CH_3$, R_2 = phytyl) and chlorophyll b (R_1 = CHO, R_2 = phytyl), and (*b*) geniposidic acid.

preventing liver cancer in adults that were exposed to the carcinogen aflatoxin (Higdon & Drake 2009).

Gardenia Blue Iridoid Pigments

The selection of natural blue food colorants for use in the United States and Europe is even more limited. In Japan, gardenia blue, which is an iridoid pigment that is extracted from *Gardenia jasminoides*, is approved for food use. In manufacturing the colorant, glucose is removed from the terpene glycoside, geniposidic acid (**Figure 4**), by enzymatic hydrolysis, and then the aglycone genipin is treated with amino acids to produce the blue pigment (Pintea 2008b, Francis 1996).

BROWN COLOR

Caramel

Brown is not a spectral color. The visual appearance is a result of multiple chromophores absorbing throughout the visible spectrum. An absorbance spectrum of caramel colorants will show decreasing absorbance from lower to higher wavelengths in the visible spectrum. Caramel is approved for general food usage in the United States, Europe, and most other countries throughout the world. Caramel colorants are produced by controlled heating of sugars with food-grade acids, alkalis, ammonium, and sulfite compounds (Sepe et al. 2008). The pigment produced is the end product of the classic Maillard reaction of reducing sugars with amino compounds. Four classes of caramel coloring are manufactured for different food applications. Their color can vary from yellow to amber to reddish brown to dark brown, depending on class and concentration. They are all water soluble, but they differ as to their colloidal charge. Class I caramel colors are produced from sugars derived from a variety of carbohydrate sources; neither ammonium nor sulfite compounds can be used as reactant ingredients. Class I caramel colorants possess a slightly negative colloidal charge. They are stable in solutions containing up to 70% ethanol and widely used in alcoholic beverages and coffee products. In production of Class II caramel colors, sulfite compounds must be used, whereas use of ammonium compounds is not permitted. They have a negative colloidal charge and are stable in alcohol up to 70%. They are used in high-proof alcoholic beverages, such as cognac, that contain tannins. In manufacturing Class III caramel colorants, ammonium compounds must be used, and sulfites are not allowed. Class III caramel colorants carry a positive colloidal charge. This prevents them from coalescing and precipitating in beer, which contains positively charged proteins. Class IV caramel colorants are produced using both ammonium and sulfite compounds as reactants. They have a negative colloidal charge, and possess good acid, alcohol, and salt stability. Class IV caramel colorants account for more than 70% of all caramel color produced. The major use is in soft drinks. Caramel colorants account for more than 90% by weight of all colors produced (Sepe et al. 2008).

Toasted Partially Defatted Cooked Cottonseed Flour

Another color additive exempt from certification (Code of Federal Regulations 2011) that can be added to foods to give colors from light to dark brown is toasted partially defatted cooked cottonseed flour. It is not accepted as a food colorant in the EU.

BLACK AND WHITE

Black is the color of objects that do not reflect or emit light from the visible spectrum. Although black is considered to be achromatic or hueless, in practice it is considered to be a color. A combination of colorants that results in light being absorbed throughout the spectrum produces a black appearance. This same appearance can be obtained with very high concentrations of individual colorants. Vegetable carbon black (E153) is used as a colorant in the EU and other countries. At one time it was an approved food colorant in the United States, but its use is no longer permitted (Francis 1999). Squid ink is traditionally used as a food ingredient for some pastas and sauces. It is considered to be part of the edible portion of squid; however, it is not legally a permitted color additive in the United States (Francis 1999).

White is the perceived color when objects have a high degree of reflectance at all wavelengths in the visible spectrum. The inorganic compound titanium dioxide (TiO_2) is a whitening agent approved for food use. Its use in the United States is limited to less than 1% by weight (Code of Federal Regulations 2011). TiO_2 is insoluble in all common solvents and available in water- and oil-dispersible preparations. It is used in confectionary products, icings, baked goods, and dairy products. There are clouding agents that are commercially available for lightening the appearance of beverages. They are emulsions formulated from a variety of food-grade ingredients.

A relatively recent addition to the listing of food colorants exempt from certification is mica-based pearlescent pigments. With this novel technology, titanium salts are deposited on mica to give an attractive, luminous visual appearance. There are different product lines that incorporate various colorants. Some appropriate applications are for hard and soft candies, chewing gums, icings, and cereals.

LOOKING AHEAD: WHAT MAY BE ON THE HORIZON?

New Sources and New Technologies

It should be emphasized that the colorants that are commercially available as alternatives to FD&C certified colorants are not stock commodities. Price is not the only variable to consider when selecting a given colorant. Colorants from different suppliers can vary in purity, tinctorial strength, shade of color, presence of unwanted flavors, tendency to precipitate, stability to heat and light, and their suitability for different applications. They need to be evaluated for their effectiveness in individual product formulations. Anthocyanin-derived colorants are a case in point. Fruit and vegetable juices are the principal source of anthocyanin colorants approved for use in the United States. As previously discussed, hue, stability, and color strength varies tremendously from one fruit or vegetable to another. The global search for edible plants with high concentrations of anthocyanins with desirable colors and good stability is an ongoing pursuit. Selection of fruit and vegetable genotypes with higher pigment concentrations is an objective of academic, government, and industry research projects. New selections may be patented or secured as proprietary assets. Several research groups and companies have investigated production of anthocyanin pigments by plant tissue culture with the view that biotechnology could be an alternative to traditional crop production (Delgado-Vargas & Paredes-López 2003). However, no food colorants produced by this technology have been commercialized. There are a wide range of unit operations and process variables encountered in fruit and vegetable juice production. There are enzymes that are used as processing aids to increase pigment recovery and improve stability. There are different filtration, microfiltration, evaporation, and resin treatment technologies that can enrich pigment content, remove flavors, and prevent sediment formation. Hence, the functionality of fruit or vegetable juice concentrates from different suppliers can vary tremendously.

More traditional extraction methods for such colorants as cochineal, annatto, paprika oleoresin, tomato-sourced lycopene, marigold-sourced lutein and zeaxanthin, and turmeric are being replaced with new technologies that use more environmentally friendly solvents and give higher yields of extracts with better color and flavor properties (Socaciu 2008c). These include supercritical fluid extraction with CO_2, pressurized liquid extraction, microwave-assisted extraction, ultrasound-assisted extraction, continuous countercurrent extraction, solid phase extraction, and microextraction. Both hydrophilic and lipid-soluble colorants can be protected from oxidation by encapsulation technologies. Spray drying, drum drying, and freeze drying are applied to formulations that incorporate the colorant with a variety of substances, including alginates, carrageenans, gum arabic, pectin, gelatin, maltodextrins, cyclodextrins, and starch. Microencapsulated lipophilic colorants and microemulsions having sizes ranging from 50 to 100 nm will be transparent and visually clear, which extends their use in aqueous systems where clarity is desired.

Regulatory Changes

Considerable energy is expended in the corporate world in attempting to anticipate changes in food colorant regulations. The possible delisting of a colorant because of new information relating to its safety, toxicity, or allergenicity can induce frustration and anxiety in the workplace. A more positive viewpoint is that increased harmonization of colorant regulations internationally may occur in the future. Other possibilities are the removal of certain restrictions. For example, extending the usage of sodium-copper chlorophyllin to additional food products in the United States is a likely possibility.

Reds

Monascus pigments that are used as a food colorant in several Asian countries are produced in China, Taiwan, and Japan (Pintea 2008b). Red yeast rice is a fermentation product produced by the fungi *Monascus* growing on rice that is traditionally used to color a variety of Asian foods and also used in traditional Chinese medicine (Mudgett 2000). The monascus pigments are soluble in lipids and ethanol, and slightly soluble in water. The colors range from orange to red to purplish-red, and are influenced by pH. They are light sensitive. Fermentation using different *Monascus* strains and different substrates can produce a range of colors from yellow to purple and also produce pigments that are water-soluble.

POP: phloridzin oxidation products

Carthamin is a red to yellow dye that is derived from safflower flowers; it has a long history of use as a textile dye (Francis 1999). The pigments are in the chalcone group of flavonoids. Carthamin is used as a food colorant in Japan. Another colorant that has been used as a textile dye is red sandalwood. The water-insoluble, red-colored santolin pigments are extracted from the heart of the Indian red sanders tree. They have been used in alternative medicine and in toothpastes, herbal mixtures, soaps, and skin care preparations (Whole Herb Company 2011).

Phycobilins are accessory photosynthetic pigments found in red and blue-green algae and cyanobacteria (Francis 1999, Pouvreau et al. 2008). They are water-soluble pigment-protein complexes. Phycoerythrin is a member of this red-colored class of pigments; it exhibits a yellow fluorescence that could be exploited in some novelty food products. There is potential for growing the algae in bioreactors and then isolating semipurified fractions by centrifugation and precipitation techniques. Spirulina is a dietary supplement produced from two species of cyanobacteria, *Arthrospira platensis* and *Arthrospira maxima*. Spirulina is a potential commercial source of the phycobilin pigments, as well as chlorophyll and several carotenoids.

Although iron oxides occur in nature, it is a bit of a stretch to consider the synthetic iron oxide that is widely used in pet food in the United States a natural colorant. Its use is restricted to pet food in the United States, but in the EU it is approved for use in foods and beverages. The water-insoluble compounds have been used for coloring salmon, shrimp, and other meat pastes.

Yellows

There are yellow or golden beets commercially grown that do not contain the red betacyanin pigments but do contain the yellow betaxanthin pigments. Because beet powder and beet juice are approved colorants, a yellow colorant from that source has been investigated (Stintzing & Carle 2008). Difficulties with browning reactions because of high levels of polyphenoloxidase activity in the beets have been a problem. Selective plant breeding to obtain yellow beets with low polyphenoloxidase activity, along with strategies to inhibit polyphenoloxidase in processing, could make this a possible source for a water-soluble yellow colorant.

French investigators have determined the structure of a water-soluble, intense yellow colorant that can be produced from apple pomace (Guyot et al. 2007, Le Guernevé et al. 2004). They have developed a process where the naturally occurring dihydrochalcone phloridzin is oxidized by polyphenoloxidase to form a stable yellow pigment, referred to as phloridzin oxidation products (POP). The scale of apple juice production is such that production of a new yellow colorant utilizing food processing wastes is an attractive possibility.

Sorghum grains have attracted interest as a potential new source of natural colorants because they contain 3-deoxyanthocyanins (Awika et al. 2004). These pigments are yellow and orange colored and water soluble and are more stable to pH-induced color change than the common anthocyanins (Ojwang & Awika 2010). Gardenia yellow as a source for the pigment crocin was discussed previously; it is approved for food use in Japan. The carthamin dyes were discussed above, and carthamus yellow (source = safflower flowers) is used as a food colorant in some Asian countries.

Greens, Blues, and the Remaining Palette

Probable extensions to approved green colorants in the United States are chlorophyll extracts or chlorophyll extracts in which magnesium has been replaced with copper or zinc. The iridoid class of pigments was previously discussed in regards to gardenia blue. Food colorants derived from gardenia fruits are particularly of interest given that not only blue, but also yellow, green, and red colorants are a possibility (Francis 1996).

The algae-derived phycobilin pigments were discussed above in reference to red-colored phycoerythrin. Algal extracts of *Porphyridium aerugineum* are blue colored, the major pigment being phycocyanobilin, which exhibits a red fluorescence.

Adulteration

Natural colorants are a high-value item. Individual colorants can come into short supply because of crop failure or political trade issues. Limited availability and high demands lead to high price, which creates an environment in which the unscrupulous cannot resist the temptation to cheat. Adulteration of natural colorants with inexpensive synthetic colorants and other dyes has occurred in the past, and undoubtedly will occur in the future. There are cases in Europe in which chili powder, paprika, and carthamus have been adulterated with Sudan red (Rayner 2007). The problem is such that the European Commission requires products to have documentation confirming the absence of Sudan dyes in foods (Di Anibal et al. 2011). Thus, in addition to price, quality specifications, and functionality, authenticity of colorants must be considered.

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