Food Coloring: The Natural Way
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Abstract
Color is a measure of quality and nutrient content of foods. The objective of adding color to foods is to make them appealing, augment the loss of color during processing, to improve the quality and also to influence the consumer to buy a product. At present, the demand for natural dyes is increasing worldwide due to the increased awareness on therapeutic and medicinal properties and their benefits among public and also because of the recognized profound toxicity of synthetic colors. Natural dyes are those derived from naturally occurring sources such as plants, insects, animals and minerals. Among all the natural dyes, plant-based pigments have medicinal values so are mostly preferred. Today the food industry and color suppliers are however constantly motivated to work towards the improvement of the technical and physical properties of the color preparations. Development of cost-effective, viable technology for the preparation of a food color and its application in foods is a challenge and the need of the day. This review article covers recent developments in technological advances of food colors with respect to natural color application and stability in foods compared to synthetic colors and the detail basic chemical information about of the major pigments.

Keywords: Natural food color, dyes, extraction, pigments.

Introduction

Why Are Color Additives Used In Foods?: The first to feast is the eyes. This is an old axiom with very significant meaning and depicts how colors in life have importance. People’s perception is usually influenced by the appearance of the food and this indicates the flavor. So it is important to note that the color of a food or beverage often dominates over other sources of information regarding the flavor. From various studies it has been observed that color of a food or beverage can play a profound role in flavor perception. Different foods are associated with different colors by people. When this perception is altered it often has a detrimental effect on the psychology of how that food taste. So color strongly influences the hospitality industry as color attracts people. Color is an important quality attribute of foods. The objective of adding color to foods is to make them appealing, augment the loss of color during processing, to improve the quality and also to influence the consumer to buy a product. Color is added to food for the following reasons: i. to replace and restore color lost during processing, ii. to enhance color that is already present, iii. to minimize batch variations in processing and iv. to color the uncolored food. Food colors can be grouped divided into four categories: a. natural colors, b. nature-identical colors, c. synthetic colors and d. inorganic colors.

History of color: The earliest written record of the use of natural dyes dates back to 2600 BC in China and addition of colorants to foods is reported in Europe during the Bronze Age. It is also reported that around 1500 BC in Egyptian cities candy makers used to add natural extracts and wine to improve the appearance. The first synthetic color (mauvine) was developed by Sir William Henry Perkin way back in 1856. The beginning of the 19th century was remarked for the bulk of production and recovery of synthetic colors from the petroleum derived products like aniline, therefore they were called ‘coal-tar’ colors because the starting materials were obtained from coal.

Color and Perception: Marketing strategy of food by major manufacturers are greatly influenced by color. Color affects almost everything one does in life is the statement given by Downham and Collins i.e., from purchasing items for one’s home, his/her clothing, or their food, decisions are made while giving color the at most importance. Their hypothesis is that all individuals are sensitive to the color of food. Appetites are also influenced and stimulated by color; color may sometimes discourage eating certain foods and diminish the desire for that food. Colors also suggest the flavors that are anticipated when eating or drinking. Bright orange colored drinks imply the flavor of orange. Similarly, a bright red colored drink may hint at strawberry or cherry or beetroot flavor. Dull colors of orange or red drinks may indicate a lesser quality of nutrients and so are not as appealing. From past 20 to 30 years many food industry are being processed to create visually appealing food that tastes good that can be used by common man. This briefs out the importance of color to food choices.

Market trends: The demand for food color in global market in 2000 was 2400 MT which increased to 3000 MT by the year 2005 and further to increase to 8000 MT by the year 2010 and is expected to increase to 15000 MT by the year 2015. The
Investment in natural food color market across the globe has touched to US $ 1 billion and is continuously growing as there is demand for natural food colors against synthetic food colors. Because of consumer’s choice for ‘natural’ food processing industry and have contributed to the increase in natural color market significantly\(^7,8\).

**Booster for Natural colors:** i. As there is increasing awareness about the harmful effects of usage of synthetic colors and the chemicals obviously demand for natural food colors in the international market abruptly increases. ii. As Japan and all European countries have banned trading of synthetic color made products. iii. As encouragement for using Natural food colors in novel products like infant toys and crayons, organic textile printing, handmade paper etc has been implemented and followed in few developed countries.

But colors from plant, animal and mineral sources also sometimes called as biocolors, which were used in earlier times, had their own drawbacks like heat, pH and light instability, and against oxidizing agents in food, which made synthetic colors gain popularity in food industry. In contrast, chemically synthesized colors were easier to produce, cinexpensive, and superior in coloring properties as they blend easily. As the use of synthetic colors in food increased, the safety concerns are also raised through numerous regulations across the world and in the USA, only seven synthetic colors are permitted. In India according to The Prevention of Food Adulteration Act of India the use of eight synthetic colors in specified food commodities at a uniform level of 100 mg kg\(^{-1}\) or mg l\(^{-1}\) is permitted\(^6,8\).

**Sources of Natural Dyestuffs:** Plant sources include roots, berries, flowers and leaves. Red color (dyer’s root from Madder plant, Brazilwood, beetroot, cranberry, safflower and orchil), orange color (stigmas of saffron flower), yellow color from (Camomile and Milkwort flowers and Weld), green color (ripe Buckthron berries, ragweed) and blue color (Woad plant and Spirulina). The most important dyes extracted from animal sources are Natural Sepia (from the ink sac of the cuttlefish), Crimson (From the Kermes Louse) and Tyrian purple (from the Murex shellfish)\(^3,9\).

**Color and Taste Perception:** The importance of color on taste perception is vital when people decide whether to eat or not. People associate different foods with different colors. In nature color = Nutrient density. According to Williams common color and food associations are noted in the table 2\(^3,9\).

<table>
<thead>
<tr>
<th>Sl.No</th>
<th>Color</th>
<th>Chemical classification</th>
<th>Plant Sources</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Orange-yellow</td>
<td>Flavone dyes, Isoquinoline dyes, Polye [dyes, Pyran dyes, Chromene dyes</td>
<td>Marigold, β-carotene, lycopene, gentisim, turmeric, Saffron, Sanguinaria Ca[ndensis</td>
</tr>
<tr>
<td>2</td>
<td>Brown</td>
<td>Napthochinone dyes</td>
<td>Camellia thea, Lawsonia inermis</td>
</tr>
<tr>
<td>3</td>
<td>Red</td>
<td>Chinone dyes, Anthrachinone dyes, Chromone dyes</td>
<td>Anatto, Beeta vulgaris, Paprica, grapes vitacea, Alkanna tinctoria</td>
</tr>
<tr>
<td>4</td>
<td>Purple-Blue</td>
<td>Benzopyrone dyes, Indigoid dyes, Indole dyes</td>
<td>Centaurea cyanus, Indigoferat inctoria Vaccinium myrtillus, Indigoferat inctoria</td>
</tr>
</tbody>
</table>

**Table-1**

**Chemical Classification of Natural Colorants**

<table>
<thead>
<tr>
<th>Sl.No</th>
<th>Color</th>
<th>Chromophore</th>
<th>Plant Sources</th>
<th>Nutrients</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Purple-blue</td>
<td>Anthocyanins</td>
<td>Eggplant, blackberry, purple, cabbage, plum, blueberry, raisins, prunes, purple grapes, figs</td>
<td>Lutein, zeaxanthin, resveratrol, vitamin C, flavanoid, ellagic acid, quecetin</td>
</tr>
<tr>
<td>2</td>
<td>Green</td>
<td>Chlorophyll</td>
<td>Avocado, cucumber, spinach, kale, broccoli, snow pea, zucchini, artichoke, lettuce, kiwi</td>
<td>Lutein, zeaxanthin, vitamin C, calcium, folate, β-carotene</td>
</tr>
<tr>
<td>3</td>
<td>White-tan</td>
<td>Anthoxanthins</td>
<td>Cauliflower, mushrooms, parsnip, potato, ginger, onions, jicama, banana, garlic, onions</td>
<td>Ancillin, Potassium, Selenium</td>
</tr>
<tr>
<td>4</td>
<td>Yellow-orange</td>
<td>Carotenoids</td>
<td>Papaya, pineapple, apricot, pumpkin, peach, peach, carrot, orange, corn</td>
<td>β-carotene, zeaxanthin, flavanoid, vitamin C, Potassium</td>
</tr>
<tr>
<td>5</td>
<td>Red</td>
<td>Lycopene or Anthocyanins</td>
<td>Cranberry, beet, watermelon, tomato, strawberry, pomegranate</td>
<td>Ellagic acid, quecetin, Hesperidin etc.</td>
</tr>
</tbody>
</table>
Chemistry of natural colorants derived from plants

Purple to blue color
Centaurea cyanus (Cornflower, figure 1a) is used for coloring sugar, confectionaries and as one of the ingredient in tea. The petals of cornflower find use in salad, cornbread muffins and also used to garnish food items. It is used traditionally for the treatment of indigestion, regulation of kidney, gall bladder and liver. Its medicinal properties include regulation of menstrual disorder, in increasing immunity and also effective in washing out wounds. It is also used in the treatment of mouth ulcers, bleeding gums and for constipation. It has antioxidant, antibacterial and astringent properties hence can be used to cure ulcers, bleeding gums and for constipation. It has antioxidant, antibacterial and astringent properties hence can be used to cure irritated or inflamed skin. It is also used in hair products and cosmetics. The distilled water from its petals is used for weak eyes and conjunctivitis. A dark blue dye can be obtained from the petals of the flower that can be used to color fabric.

Indigofera tinctoria: Figure 1b is used to cure constipation, liver disease, heart palpitation and gout. Coloring matter (Indigotin) is usually present abundantly in flowers which give blue color mainly used to dye linen and hairs. Flavonoids, terpinoids, alkaloids, glycosides, Indigotine, Indirubin, rotenoids are the other related compounds abundantly present in the plant. These compounds were found to be responsible for many pharmacological activities such as anti-hyperglycemic activity, antioxidant, anti-inflammatory activity, antibacterial, antihepatoprotective activity, antidiabetic activity and anticonvulsive agent.

Vaccinium myrtillus: Bilberry, figure 1c the fruit juice is red in color and this turns blue in basic medium. The extract can be used for treating bladder stones, biliary disorders, scurvy, coughs, and lung tuberculosis. More recently, bilberry fruit extracts have been used for the treatment of diarrhea, dysentery, and mouth and throat inflammations. Bilberry leaf decoctions have been used to lower blood sugar in diabetic patients.

Sambucus nigra: Blueberry, figure 1d are edible berries. Flowers are used for medicine, fruits as dyes for basketry, arrow shafts, flute, whistles, clapper sticks, and as folk medicine. The active alkaloids in elderberry plants are hydrocyanic acid and sambucine. Both alkaloids will cause nausea so care should be observed with this plant. Elderberries are high in vitamin C. The red berries of other species are toxic and should not be ingested.

Red color
Annatto (figure 2a): Seeds of annatto are used for coloring Gloucester cheese since the 16th century, following with Cheshire, Red Leicester cheese and cheddar made in Scotland. In Spanish it is called as local saffron. In the European Union, annatto has been given the E number E160b whereas in the United States, annatto extract is listed as a color additive “exempt from certification” which is informally considered to be a natural coloring. The yellow to orange color is due to the chemical compounds bixin and norbixin, which come under apocarotenoid. The fat soluble color in the crude extract is called bixin, which can then be saponified into water soluble norbixin.
**Beeta vulgaris:** Betanines or betalains are natural dyes extracted from different Beeta vulgaris. They are largely used as food colorants in food products like yogurts, ice cream and other products. Recent studies have shown that betanines have antioxidant, antimicrobial and antiviral activity besides betanine, another pigment which is extracted from beetroot is vulgaxanthine¹⁶ (figure 2b).

**Paprica:** The pigments of importance present in paprika are a mixture of capsanthin and capsorubin, both are carotenoids, responsible for the red color of the dye. This dye is used in cosmetics and in medicine¹⁷ (figure 2c).

**Grapes vitacea:** The red dye obtained from the grape fruit was used in history by the Kiowa to paint skin and feathers. The red to purple color is due to the presence of anthocyanins. Other valuable nutrients present are glucose, fructose, potassium, calcium, tartaric acid, malic acid, tannins and anthocyanins. The medicinal benefits of grape are it is used in preventing diseases of the heart and blood vessels, varicose veins, hemorrhoids, “hardening of the arteries” (atherosclerosis), high blood pressure, swelling after injury or surgery, heart attack, and stroke. Grape seed is also useful in diabetes complications such as nerve and eye problems, improving wound healing, preventing tooth decay, preventing cancer, in an eye disease called age-related macular degeneration (AMD), poor night vision, liver disorders, and hay fever¹⁴ (figure 2d).

**Alkanna tinctoria:** Alkanna (figure 2e) is an astringent and a source of red pigment used in cosmetics. It was traditionally used topically for the treatment of skin wounds and diseases. Orally, alkanna root has been used for diarrhea and gastric ulcers. Alkanna root has demonstrated radical scavenging activity, suggesting potential antiaging effects. Alkanna root contains a mixture of red pigments found in the bark at levels of up to 5% to 6%. These consist mainly of fat-soluble naphthazarin (5, 8-dihydroxy-1, 4-naphthaquinone) components, such as alkannin and related esters. The red pigments are soluble in fatty oils, which make them useful for the detection of oily materials in microscopic powders during histological examination¹⁴.
Brown color

**Camellia thea** (Tea, figure 3a): Black to brown color from extracted Camellia thea is used in the cosmetic industry as stated by a Japanese Chemical (Raw Material for Cosmetics) DAITO KASEI KOGYO company. Other benefits of tea include weight loss and obesity management, prevention of cancer, dental hygiene as it is rich in antioxidants.\(^{18,19}\)

**Lawsonia inermis** (Henna, figure 3b): The constituents of Lawsonia inermis include essential oils like 1,4 naphthoquinone and 5-10% tannins, gallic acid, flavonoids, lipids, sugars, triacontyl tridecanoate, mannitol, xanthones, coumarins (5-alkyloxy 7-hydroxycoumarin), 2-3% resins, and up to 2% Lawsone (2-hydroxy-1,4-naphthoquinone). Among these Lawsone (2-hydroxy-1,4-naphthoquinone) is the coloring agent. The European Commission of Health & Consumer Protection Directorate-General in 2005 has not approved Henna as a food color. This was agreed by FDA (U.S Food and Drug Administration) in 2006 because of mild toxicity of Lawsone.\(^{18,20,21}\)

**Curcuma longa** (Turmeric, figure 4a): Is one of the most essential spice used all over the world. It is called “the golden spice of life”. Since Harappan civilization the use of turmeric has been evidenced. It has been considered has the poor man's saffron because it offers a yellow coloring cost effectively. It is used as an alternative to saffron. Curcumin is the primary pigment of color. It is generally used in various food industries for coloring. Mainly used in dairy products, beverages, cereal, pickels, sausages, confectionaries, ice cream, bakery and savory products. Apart from coloring, it is also used in skin care and hair care cosmetic products as it is antibacterial in nature. It is also used in Ayurvedic medicine as analgesic, anti-inflammatory, antitumor, antiallergic, antioxidant, antiseptic, in treating anemia, diabetis, indigestion, gallstones, food poisoning, poor blood circulation.\(^{14}\)
Sanguinaria Canadensis (Blood root, figure 4b): Bloodroot has been used historically by some Native American tribes as a medicinal agent to stimulate the digestive system and induce vomiting. It has also been used as an antimicrobial agent. Recently, the main active constituent of bloodroot; Sanguinarine, has been added to dentifrices (used to clean teeth) to reduce plaque and treat gingivitis and periodontal disease as reported in 2003 from the U.S. Food and Drug Administration (FDA) Dental Plaque Subcommittee of the Nonprescription Drugs Advisory Committee. They also concluded “that sanguinaria extract at 0.03-0.075% concentration is safe. It is also used to empty the bowels, to treat cough, hoarseness (laryngitis), sore throat (pharyngitis), poor circulation in the surface blood vessels, nasal polyps, achy joints and muscles (rheumatism), warts, and fever\footnote{18,22}.

Tagetes erecta (Mexican Marigold, figure 4c): Lutein from Tagetes erecta L. is a purified extract obtained from marigold oleoresin. Lutein is extracted from the petals of marigold flowers with organic solvents which impart yellow to orange color. It is used as a food coloring agent and nutrient supplement (food additive) in a wide range of baked goods, beverages, breakfast cereals, chewing gum, dairy product analogs, egg products, fats and oils, sauces, infant and toddler foods, in levels ranging from 2 to 330 mg/kg. It is also used as antiseptic\footnote{23}.

Green color: Chlorophyll is a green pigment found in most plants, algae, and cyanobacteria. The name is derived from the Greek words (chloros "green") and (phyllon "leaf"). Chlorophyll was first isolated by Joseph Bienaimé Caventou and Pierre Joseph Pelletier in 1817\footnote{21, 3}.

Chemical structure-Chlorophyll is a green pigment, which is structurally similar to and produced through the same metabolic pathway as other porphyrin pigments such as heme. At the center of the ring a magnesium ion is present. The chlorin ring is connected to different side chains, usually a long phytol chain. There are different forms; the most widely distributed form in terrestrial plants is chlorophyll a. The general structure of chlorophyll a was elucidated by Hans Fischer in 1940, and in 1960, Robert Burns Woodward published a total synthesis of the molecule. In 1967, the last remaining stereochemical elucidation was completed by Ian Fleming, and in 1990 Woodward and co-authors published an updated synthesis. Chlorophylls, a group of fat soluble natural pigments, are obtained by solvent extraction of grass material, lucerne and nettle. The principal coloring matters are the phaeophytins and magnesium chlorophylls, which are highly unstable to light. The green color is due to the pigments chlorophyll a (blue-green) and chlorophyll b (yellow-green) that occur together in a ratio of about 3:1.22 Chlorophyll is converted to chlorophyllins in presence of alkali, which renders it water soluble. The technological advance in chlorophyll pigment is that the magnesium atom in the structure is replaced by zinc or copper, which improves its stability to light\footnote{21, 3}.
chlorophyll a          chlorophyll b
structure of chlorophyll pigment

chemistry of natural colorants derived from animals

Dactylopius coccus (Cochineal, figure 5a): is native insect of South America and Mexico. It is a parasite which lives on cacti of genus Opuntia feeding on moisture and nutrients. The dye stuff extracted from this insect and its eggs is Carminic acid (Carmine), which is red in color. Carmine is used as a food dye in juices, ice cream, yogurt, and candy, and as a dye in cosmetic products such as eye shadow and lipstick. But as a food dye it has been known to cause severe allergic reactions and anaphylactic shock in some people.

Sepia officinalis L (Female Cuttlefish, Figure 5b): it has rich concentrates of orange-red pigment in the accessory nidamental glands. The pigment is called Sepiaxanthine. The dye is called Sepia ink. The sepia pigment is used in capsule printing ink which has been patented in European Patent Application EP1361258. It is used in Spanish cuisine breaded and deep-fried cuttlefish is a popular dish in Andalusia. In Portugal Chocos com tinta is served as deep-fried strips 'cuttlefish in black ink'.

Black pasta is often made using cuttlefish ink. The ink is a mixture containing melanin, protein, carbohydrate and lipid. It is reported that protein constitutes approximately 10.08% of squid ink from Sepia latimanus. Additionally, the dark natural material contains nine fatty acids (approximately 1.34%) of which 43.4% are unsaturated fatty acid and 56.6% is saturated fatty acid plus sixteen amino acids of which aspartic acid is the richest.

Cephalopod is another member of the molluscan class Cephalopoda, Figure 5c: Cephalopod ink is generally obtainable from fishmongers or gourmet food suppliers and in cooking, it is used as a food coloring and flavoring, in pasta and sauces.

Monascus purpureus (figure 5d) - Monascus purpureus are fungus. The red pigments produced by this fungus were traditionally used in oriental countries, because of its potential application as food additives. The use of this color additive is not yet regulated in the European Union, United States and Brazil, Philippines, Taiwan among other regions. Oriental countries such as Japan make extensive use of these pigments since decades - as water soluble pigments in candies (Watanabe, 1997), or red pigment for red rice wine. It is also used as cholesterol-lowering agent.
Figure-5
Pigments that are derived from animals [a] Dactylop ius coccus (Cochineal), [b] Sepia Officinalis, [c] Cephalopod and [d] Monascus purpureus

Methods of Extraction of Pigments

Solvent extraction is the conventional method that is usually followed to extract colors from plant materials. Anthocyanin and betalain pigments, which are water soluble, are extracted from the raw material with water and sometimes with aqueous methanol. For carotenoids extraction, hexane is the solvent of choice and acetone is good choice of solvent for the initial extraction of pigment from the plant material. After thorough extraction of the plant material, the extract is concentrated and subjected to purification steps by using column chromatography. Identification and quantification of the pigment is performed by spectrophotometry or by high pressure liquid chromatography (HPLC)\(^2\).

The advancements in extraction of pigments from plant materials were necessary as the use of organic solvents is harmful both for health as well as for environment. The current advance techniques that are followed in color extraction are as follows: i. High Hydrostatic Pressure (HHP), ii. Pulsed Electric Field (PEF), iii. Sonication-assisted Extraction, iv. Gamma Irradiation, v. Enzymatic Extraction, vi. Membrane Technology.

High Hydrostatic Pressure (HHP) and Pulsed Electric Field (PEF)

High hydrostatic pressure (HHP), pulsed electric fields (PEF) and sonication belong to environment friendly category and energy efficient technologies that enhance mass transfer processes within plant or animal cellular tissues, as the permeability of cytoplasmatic membranes can be increased which in turn enhances extraction of valuable cell components. In addition, the decrease in the dielectric constant of water under HHP combined with temperature leads to a decrease in the polarity of the media, contributing to the higher yield of total phenolics and other antioxidants. PEF is reported to enhance mass transfer rates by electroporation of plant cell membranes, improving tissue softness and thus influencing the textural properties. PEF is reported to be an ideal method to enhance juice production, increase the extraction of valuable components better than the yields obtained by enzymatic maceration\(^2,30\).

Sonication-assisted Extraction

Sonication is one of the most commonly used methods to enhance mass transfer phenomena by cavitation forces, where bubbles in the liquid/solid extraction can explosively collapse and generate localized pressure, causing plant tissue rupture and improving the release of intracellular substances into the solvent. Its feasibility for the extraction of secondary metabolites such as tea, mint, chamomile and ginseng has been highlighted in many studies. A few studies on application of sonication to extraction of food colors have been reported\(^2,31\).

Gamma Irradiation: Gamma-irradiation, as a pre-treatment to a plant material, increases cell wall permeabilization, resulting in enhanced extraction of cell constituents in higher yield\(^2,32\).

Enzymatic Extraction: Enzyme assisted extraction of worthy products from plant materials viz., pigments, antioxidants, flavors and phytochemicals is another new technology. Enzyme pretreatment cannot be a complete substitute for conventional solvent extraction, but can result in increased yield of value added cell components and a reduction in time of extraction and reduction in amount of solvent consumption. Based on this approach, enzymes have been explored as a means to enhance the extraction of carotenoids in marigold flowers\(^2,33\).

Membrane Technology: Membrane processing is a fast and emerging technique for the concentration and separation of macro and micro molecules based on molecular size and shape in biotechnology and food processing industries. Advantages of membrane processing are many which include improved product quality with higher yield, utilization of byproducts, temperature and pH sensitive products can easily be extracted without alteration and lastly is environmental friendly as no harmful chemicals are being used and less energy is consumed\(^2,34,35\).

Future Perspectives: The choice of appropriate color to food is very challenging and wrong selection of color may lead to lot of problems viz., lack of consumer appeal and the failure of product. Color suppliers are facing challenges of producing cost effective, good stability, easy to handle, technology for the
production of colors. In this attempt, new technologies are welcome with improved benefits over existing technologies.2,3,8

Conclusion

Nowadays, natural dyes and colors have growing importance not only in dyeing but also because of their medicinal properties. As the awareness among people towards natural dyes and their therapeutic uses are increasing because of their non-toxic or less toxic properties, fewer side effects. To meet the need and demand of growing population, emerging technologies such as high hydrostatic pressure, pulsed electric fields, sonication and membrane technologies could be potential methods for the enhanced extraction of pigments and bioactive compounds from plant or animal materials in future. Our concern towards environment is also increasing day by day, so keeping that in mind it can be easily anticipated that membrane processes could be the technology of the future in the food and allied industries. The importance of understanding the effect of color on food choices and flavor perception is of paramount to hospitality industry employees as well. To conclude, there is need for proper methods for extraction, documentation and characterization of dye yielding plants and animals and dye pigments obtained for further development of pharmaceutical industry to formulate the natural, safe, environmentally friendly plant and animal pigments into therapeutically beneficial pharmaceutical formulations/dosage forms for safe use.

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