

Review on Natural Colorants Used in Cosmetic

Rajlaxmi Deolekar¹, Vaibhav R. Urade², Bilal Sufi³, Aishwarya Shirrao⁴

New Montfort Institute of Pharmacy, Ashti, Wardha, Maharashtra^{1,2,3,4}

vaibhavurade10@gmail.com

Abstract: This review offers an examination of the properties and recent advancements in research concerning the utilization of plant-derived colorants across the domains of food, cosmetics, and textile materials. The comprehensive analysis encompasses various types of colorants, including polyphenols such as anthocyanins, flavonol-querceetin, and curcumin, as well as isoprenoids like iridoids, carotenoids, and quinones. Additionally, N-heterocyclic compounds such as betalains and indigoids, along with melanins and tetrapyrroles, are scrutinized for their potential industrial applications. The review also delves into future perspectives, exploring the evolving landscape of applying plant-derived colorants in the coloration of diverse materials.

Keywords: Plant-based Colorants, Anthocyanins, Isoprenoids, Betalains, Cosmetic Industry, Textile Applications, Food Coloration

I. INTRODUCTION

There is presently a resurgence in the adoption of natural ingredients evident across diverse facets of human life. This revival extends beyond phytotherapy and encompasses the imperative to craft products utilizing natural raw materials, prominently including plant-derived ingredients. Industries are progressively embracing ecological practices, prioritizing environmental sustainability and prioritizing consumer health. The widespread use of various herbs, vegetable oils or essential oils in various products is remarkable. In this paradigm, a key category of compounds emerges - colourants, which play an essential role in many applications. While the textile industry presently dominates colorant consumption, their application extends to diverse sectors such as paper, leather, printing, plastics and chemistry indicators.

The colorants market is a highly specialized sector, often considered closed, yet it is significantly influenced by consumer preferences for natural over synthetic alternatives. Consequently, the global market for natural dyes is expanding at a faster rate than the global dye market, primarily propelled by their increasing utilization in the food industry. Presently, in the textile industry, plant-derived dyes are mainly used for in craft dyeing. In the cosmetics sector, the EU's list of dyes and pigments, comprising 153 compounds, reveals that approximately 8% are derived from plants, 20 percent are inorganic dyes, while the majority—82%—comprises synthetic dyes and pigments. Notably, among plant-derived dyes approved for cosmetic use based on applicable regulations are β -carotene, canthaxanthin, anato, lycopene, curcumin, vegetable carbon, riboflavin, caramel, pepper extract, captin, beetroot red, and various anthocyanides (cyanides, peonides, malvidin, delphinides, petunides, pelargonides).

The historical precedence of plant-derived dyes in industries like textiles, leather, paper, inks and art highlights their traditional significance. However, the advent of synthetic colorants, starting with mauve in 1856, marked a shift in the dyeing industry. Synthetic alternatives gradually replaced natural colorants obtained from plants and animals, with subsequent breakthroughs leading to the creation of organic pigments. While synthetic colorants offered a broader spectrum and simplified manufacturing processes, recent societal trends favor a return to natural ingredients.

Polyphenol Applications:

Anthocyanins:

Anthocyanins, a prominent subgroup of flavonoids, are chiefly responsible for imparting color to various plant sources. Rich anthocyanin content is found in diverse plants such as blackcurrant, blueberries, raspberries, wild rose, elderberries, red grapes, chokeberries, blackberries, cornflowers and black corn. Additionally flowers of the hibiscus, mallow petals and leaves of red cabbage, onions or root berries also house these pigments. Anthocyanins are

omnipresent in fruits, flowers, leaves and storage organs, with concentrations ranging from 0.1% to 1% dry weight in certain plants. Traditional methods of extracting anthocyanins involve liquid–liquid extraction using organic solvents, albeit these can be toxic and environmentally unfriendly. Contemporary extraction techniques include supercritical fluid CO₂ extraction and ultrasound extraction and the application of hydrolysis enzymes.

Anthocyanins are characterized by their sensitivity to light, oxygen, and temperature, necessitating storage in low temperatures, devoid of light and with limited oxygen availability. To address this, short-term strategies like copigmentation and self-association, as well as long-term approaches such as creating polymeric colorants or pyranoanthocyanins, have been proposed. Pyranoanthocyanins, identified in red wine, offer enhanced stability and color longevity compared to traditional anthocyanins.

While anthocyanins are commonly food dyes are often used as red dyes limits industrial applications. Studies have explored co_pigmentation and encapsulation processes to enhance colour stability and antioxidant activity. Research has also identified stable anthocyanins under less acidic conditions, broadening their potential applications and various food products. Sources like black carrot, Cornelian cherries, radish, eggplant, and others present promising options for stable anthocyanin-based natural colorants in the food industry.

Flavonols-Quercetin

A specific member of the group of flavonol compound, quercetin, exhibits notable potential for dyeing textiles, particularly cotton, in various colors. A research study by Adeel et al., both irradiated and un-irradiated plant powder were subjected to UV radiation along with textiles. Mordants such as tannic acid, copper sulphate, iron sulphate and aluminum sulphate have been introduced and the colour under friction, washing and light exposure to understand the effect of UV radiation. The optimal conditions for satisfactory colorfastness were identified as 50 minutes of dyeing at 55 °C, pH 6 and iron salt salts 8 g/100 ml.

Additional studies have explored the application of quercetin from to colour cotton fabrics, onion shell powder. Similar in previous research, dye was extracted from irradiated and un-irradiated onion shell powders and the resulting solution was used for dyeing irradiated cotton fabrics. To enhance color stability and strength, iron and aluminum used before and after mortar. The optimal conditions for achieving satisfying color performance and maximum strength include 60 °C temperature, 1:30 alcohol ratio, 10% per-mordant alkaline and 6% post-mordant alkaline concentration. Rehman et al. and colleagues that gamma radiation enhanced the colourfastness and colour strength of dyed cotton.

Curcumin-Turmeric:

Turmeric rhizome is valued as both a spices plant and as a dye used in the manufacture of food and cosmetic. The yellow hue attributed to turmeric is the result of curcumin, demethoxycurcumin, and bisdemethoxycurcumin. Curcumin, in particular, turmeric ground was the subject of a demonstration by the American University of Technology in the United kingdom maintains stability at moderate temperatures, allowing for a storage period of almost six months, although susceptibility to microbiological hazards exists. While water activity has a limited impact on stability, degradation occurs in the alkaline environment and turmeric is light sensitive.

In a study comparing curcumin and tartrazine stability in an extruded food product made from soybeans and corn flour, curcumin demonstrated a faster degradation rate. However, the authors suggested its potential to be an alternative to synthetic dyes. Turmeric also finds application in textile dyeing, acting as both a colorant and biomordant. Cotton fabrics treated with turmeric biomordant and coloured with black carrot leaf extract, the color strength was higher, attributed. In addition, the hydrogen bonds of the cellulose fabric hydroxyl group and anthocyanin, which are bound to biomordant, form additional hydrogen bonds. The optimal concentration for pre-mordanting was 2%, while post-mordanting achieved optimal results at 8%. Washing, light exposure, and heat treatment of the dyed textile yielded acceptable results.

Isoprenoid Applications:

Terpenoids, also referred to as isoprenoids, are categorized the main three groups are: iridoids, carotenoids and quinones. According to the Scopus® database, the last decade has seen the publication of approximately 621 papers featuring the term "isoprenoid" in the title and 5341 when extended to title, abstract and keywords. Despite the

collective term "isoprenoid" being the focus of scientific inquiry, it is crucial to delve into the distinctive characteristics of its subgroups, namely iridoids, carotenoids, and quinones.

Iridoids:

Iridoids are predominantly found in dicotyledonous plants, with *Gardenia jasminoides* standing out as one of the most popular sources. The ripe fruits of this plant have gained widespread use as a natural colorant, offering blue, yellow and red pigments that have been successful for many years, textiles, cosmetics and have been involved in the textiles, cosmetics and food industries. The main glucoside of iridoids obtained from *Gardenia* jasmine is geniposide. When geniposidic acid reacts with amino acids, such as glutamate and arginine, there is citric acid, it produces purple-red polymer. This polymer is water-soluble and stable at a wide range of pH, but sensitive to light. Greater light resistance is achieved with polymers containing geniposide and glycine. The aglycon part of geniposide, genipin, can be obtained by reacting with β -glycosidases, and its reaction with amino acids results in a blue pigment. Cho et al. conducted studies optimizing the production parameters for the one-step chemoenzymatic conversion of geniposide into a blue pigment, with tyrosine and glycine yielding the highest production yields. The molar ratio of genipin to amino acids that proved most optimal was unity. The resulting blue pigments were successfully utilized to dye fabrics like wool, silk and cotton, exhibiting good colorfastness even without the application of mordants.

Carotenoids:

Carotenoids, synthesized through an isoprenoid pathway involving eight isoprenoid units, are integral compounds in the realm of terpenoids. The nomenclature of carotenoids often reflects the plant from which they were initially isolated, exemplified by the first isolation of β -carotene from carrots in 1831. Responsible for a vibrant spectrum of colors including pink, red, yellow, and orange, carotenoids are ubiquitous in nature, existing in nearly every organism. In plants, they are predominantly found in plastids within leaves, fruits, and flowers, with a rare occurrence in roots. The concentration of carotenoids in plants typically ranges from approximately 0.07% to 0.2% of the dry mass, with free-form presence exclusive to leaves; in other tissues, they are esterified or form noncovalent complexes with proteins. Soluble in nonpolar solvents, carotenoids, also known as isoprene lipids, are found in various sources such as tomatoes, annatto, carrots, peaches, pumpkins, red palm oil, paprika, saffron, and wild roses. Their versatile utility extends to textiles, food, and cosmetics, including applications in self-tanning products.

Annatto:

Annatto stands out as one of the most widely utilized sources of carotenoids, containing norbixin and bixin, available in water soluble extract, oil-soluble extracts, or oil suspension forms. Renowned for imparting a red or orange hue, annatto colorant is known for its overall stability, although its color can be influenced by pH levels. Throughout history, annatto has been a prominent food colorant, particularly in dairy products and cosmetics, pharmaceuticals and textile. including materials such as silk, wool, and cotton. Gulrajani et al. conducted studies on Polyester and nylon fibers dyed with annatto, revealing the compatibility of synthetic fibers with the colorant. At high temperatures, dye absorption increases due to an endothermic process. The resulting color displayed resistance to washing but proved light-sensitive. Furthermore, distinct mechanisms were identified for dyeing ionic and non-ionic textiles. Annatto has also found favor as a colorant in cosmetic products. Aher and Bairagi incorporated varying The amount of annatto extracts into the lipstick formula, resulting in shades ranging from yellow red, to light red and dark red.

Marigold:

Marigold, an annual herb boasting lutein, serves as another valuable carotenoid source, with its dried flower petals finding application in dyeing. Recognized for its production heat - stable and light-stable dyes with high tinctorial power, marigold dye can be seamlessly blended with substances like vegetable oil, gelatin, or calcium silicate. Additionally, the cost-effectiveness of marigold positions it favorably against alternative yellow and orange dyes such as saffron and turmeric. The proper storage of marigold flowers is imperative to maintain colour stability, and innovation technology can influence this factor, techniques like anaerobic and lactic acid treatment. Extracts from marigold are employed in poultry feed, to improve the colour of the yolks of eggs. In a study conducted by Sowbhagya

et al., dried marigold petals were incorporated into butter creams, yielding promising results by enhancing the shelf life and overall quality of the food tested. The most favorable dyeing outcomes were observed with ferrous sulfate as a mordant through the post-mordanting method, with the study indicating that marigold colorant did not compromise the structural integrity of the fibers.

Saffron:

Saffron, derived from the dry *Crocus sativus* stigmas are grown in countries such as Iran, India and Spain. It boasts a rich historical legacy of use. Its primary active compounds include safranal, crocin, and picrocrocin. Safranal contributes to saffron's distinctive aroma, while picrocrocin imparts a bitter taste. The carotenoid crocin is responsible for the saffron's yellow-orange color, responsible for the beneficial characteristics of saffron of easy water solubility, a feature not shared by many other carotenoids that often require specialized formulations for aqueous solubility or dispersibility. Saffron has also been employed as a cosmetic colorant, notably in lipstick. With the current trend favoring the incorporation of natural raw materials in cosmetics, they have the potential for the revival of saffron in cosmetic applications, although limited quantities due to their high cost. Saffron can serve as an alternative to turmeric or even a botanical substitute for synthetic colorants like tartrazine. In a study by Elmaaty and colleagues, they used saffron and curcumin to dye synthetic fabrics, with pre-treatment involving UV/Ozone exposure for varying durations, resulting in an improved dyeing process for both colorants.

Paprika:

Sweet paprika, derived from the dried pods, serves as another valuable source of carotenoids, presenting as a deep red powder rich in capsanthin and capsorubin. In the food industry, paprika oleoresins extracted from this source find significant utility. Santos et al. conducted an evaluation of the properties of saline paprika oil in rice starch/gel and Arabic gum for food applications. The study revealed a satisfactory outcome, showcasing the positive impact without compromising the texture, taste, or flavor of the food product.

Tomato:

Tomatoes serve as another notable carotenoid source, housing lycopene and β -carotene. In a study by Yaldiz and Baysal, it was demonstrated that electroporation, when applied during the preheating process, could effectively boost the lycopene content in tomato puree, thereby enhancing its color. This innovative technique yielded superior color results compared to conventional steam methods.

Improving the Properties of Carotenoid Colorants:

Carotenoids exhibit sensitivity to factors such as light, temperature, pH, and redox factors. A viable approach to enhance its properties involves the microencapsulation of carotenoids, such as β -carotene, leading to prolonged shelf life and color modification. In a particular study, β -carotene is distributed uniformly in the shellac polymer matrix to reduce degradation over time, observing the resulting color changes at different concentrations of the β -carotene loaded. The addition of antioxidants, such as caffeine or propyl gallate, further improved the stability of β -carotene. Given that several natural carotenoids are water-soluble and crocin is a type of carotene, a prominent example, numerous attempts have been made to increase the number of hydrophilic carotenes. For example, the presence of a carboxyl group in Crocetin and Bixin contributes to this slight water solubility. In some cases, water-soluble powders may require ingredients such as acacia gum and modified starch. Improving the hydrophilic properties of some carotenoids may offer additional benefits, such as an increase in antioxidant activity compared to their hydrophobic counterparts in organic solvents. Stability is also a key consideration in examining carotenoids. Haas et al. evaluated carrot concentrate used to produce crystals containing crystalline carotenoids for stability during drying and storage. Various stabilization methods, including oxygen-free storage, freezing and spray drying, as well as the addition of various additives (sodium arabic, maltodextrin, modified starch, sodium ascorbate and tocopherol) were tested. All methods demonstrated loss of carotenoid, with a reduction of less than 5 percent in the processing phase. These conclusions may enhance the industrial application of carotenoids and make them more feasible for mass production.

Quinones:

The spectrum of Quinones ranges spans a broad range from deep purple to orange and yellow, making Quinones historically significant as colorant. The well-known Quinones are Alizarin, Ruby, Xantopurpurin, Purpurin, Alkanna, chrysoarobin and Cochineal. Naphthoquinone-lawsone from *Lawsonia alba* is widely used in cosmetic for coloring, while shikonin, another naphthoquinone, which was first derived from the cell culture of *Lithospermum erythrorhizon* in 1985 and was used in the Japanese cosmetic company Kanebo for the production of lipstick, creating considerable interest in the cosmetics market. Walnut extract, combined with iron sulphate, aloe extracts and ascorbic acid were used by Mital and others to colour hair and produced dark brown dyed hair that showed light resistance to and washing resistance, with acceptable color strength and no irritation.

Anthraquinones, such as alizarin and purpurin are present in the roots of *Rubia tinctorum*, which used for dyetextiles like cotton and linen. The colour of cochineal depends not solely dependent on the amount of carmine acid, but is an indicator of the quality of colorants. Chanel collaborated with Rochefort CRITT Horticoletto develop an insoluble coloring substance, selecting sappan wood with a kaolin substrate. This research led to the production a red pigments for lipsticks, gloss, eyeshadows, and blushes, based on sapon wood extracts on a kaolin substrate. Anthraquinones are not exclusive to plants and animals; they are also present in marine-derived fungi. Typically biosynthesized by one of two known pathways, anthraquinones, such as *Monascus* fungi colorants, are applied in various dyeing industries, food, textile and cosmetic sectors owing to their UV absorption properties.

Applications of N-Heterocyclic Compounds:

Indigoid:

The N-heterocyclic group of compound features indigoids, which are one of the oldest known colors in humans, renowned for dyeing textiles, particularly wool. Derived from plants such as *Indigofera tinctoria* and *Isatis tinctoria*, *indigofera* stands out as a colorant that doesn't require mordants. Although it was historically employed on a larger scale, its use has shifted to more localized applications, particularly in craft dyeing scenarios.

Betalains:

Betalains, derivatives of betalamic acid, encompass two distinct groups: the Purple-red betacyanins and yellow betaxanthins are described in more than 50 betalains in various plants. Present in fruit, flowers, stems, roots, bracts and certain higher fungi within 13 Caryophyllales families, the source of food include red beetroots, dragon fruit, cacti fruit, amaranth and radish. Betalains exhibit high tinctorial capacity and antioxidant activity, with betacyanins showing superior antiradical properties compared to betaxanthins. Generally more stable than anthocyanins, betalains endure a broader pH range and renew after thermal treatment. However, its stability is influenced by factors such as pH extremes, high water activity, metal cations, light and elevated temperatures. Acylated betalains demonstrate enhanced stability compared to nonacylated variants due to their resistance to racemization.

Since the early 20th century, betalains have served as food colorants, with applications in the cosmetic and pharmaceutical industries. Betalains from beetroots, whether in juice or powder form, are utilized in products like jams, fruit yogurts, and ice creams. Research efforts, such as those by Azwanida et al., explore the potential of betalains as colorants in cosmetics, aiming to offer alternatives to synthetic options. Dragon fruit extract-derived betalain pigments were incorporated into lipstick formulas, demonstrating stability at room temperature. However, exposure to elevated temperatures led to a darker color. Numerous studies focus on enhancing betalains' potential for industrial use, exploring methods like encapsulation with substances such as ascorbic acid, isoascorbic acid, or gluconic acid to improve color stability. Blending betalains with compounds like α -tocopherol and incorporating them into liposomes presents additional avenues for enhancing their properties.

Melanin Applications:

Melanin, another naturally occurring colorant, comprises polymers they are formed by phenolic or ethylene monomers and can complex with proteins or carbohydrates, contributing to brown, grey and black colors in animals and plants. Chestnut shells contain about 15% of plant melanin, which is a by-product of the food industry and can be used as anti-oxidative food dye. While not vegetable origin, it is noteworthy that the ink from cuttlefish *Sepia officinalis* is blacked

by melanin and has historically employed by humans used for various purposes by humans including food dyeing. The black ink from *Sepia officinalis* has found application in cosmetics, as explored by Neifar et al. has explored in mascara and shadows. The study reported favorable outcomes, demonstrating that incorporating this black ink in iron oxide and black bone dye formulations enhanced spreadability, coverage and color of cosmetics.

Tetrapyrrole Applications:

The tetrapyrrole structure can manifest as either linear (exemplified by the basic bilin structure) or cyclic (represented by the basic porphyrin ring). Plant bilins including phytochrome, phycoerythrin and phycocyanin, stand out as some of the most common pigments on Earth. Derived from photosynthetic organisms, Phycobilins and Phycobiliproteins find application as food color. For example, the phycobilin sourced from spirulina have gained approval as a colorant for various food items, such as chewing gum, candies, and dairy products, serving as almost the practically the sole plant-derived alternatives for blue and green colors. Phycobiliproteins have found widespread use, with numerous patents exploring their applications in the food, cosmetic, and even fluorescence dye industries. These stable colorants are influenced by factors such as type of protein, concentration, purification methods, temperature and pH. Notably, unlike anthocyanins, alterations in pH do not induce color changes in phycobiliproteins. Phycoerythrin, a water-soluble pink dye, present fascinating ingredients for applications in cosmetics, textiles, and pharmaceuticals. Studies on the stability of these colorants extracted from *Porphyridium* revealed resilience at temperatures around 60 °C and pH levels of 4, 5 and 6, with stability periods ranging from 30 to 40 minutes. In a comparative study on the resistance of blue food colorants to light and heat, including phycocyanin, indigo, and gardenia blue, phycocyanin emerged as the most versatile despite exhibiting lower stability under light and heat influences.

II. CONCLUSION

In conclusion, this review highlights the versatile applications and growing significance of plant-derived colorants in the food, cosmetic, and textile industries. With a focus on sustainability, health consciousness, and consumer preferences, these natural pigments offer a promising alternative to synthetic counterparts. The interplay of traditional wisdom, scientific advancements, and industrial innovation underscores the dynamic potential of plant-based colorants. As industries continue to embrace eco-friendly and health-oriented solutions, the utilization of botanical pigments is poised to shape the future of product development in these sectors.

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