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NATURAL COLORANTS FROM PLANTS FOR WELLNESS INDUSTRY

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ABSTRACT: Colorants are widely used in food, cosmetics, pharmaceutical, nutraceutical and textile industry to improve customer perception and acceptability. The hazardous effects and legislation of synthetic dyes minimize its applications thus the search for natural pigments is in trend. Plant natural pigments are widely used as alternative not only because of their high availability as colouring agent, but also they exhibits abundance of health-promoting values such as antioxidant and antimicrobial activity which favour human beings. Drawbacks of natural pigments including its stability and cost hindered its popularity in industry but the emerging of modern technologies is expected to overcome this problems. This review focuses on the natural colorant from plants source, production technologies and their implication in wellness industry; *i.e.* food, cosmetic and pharmaceutical.

INTRODUCTION: Colours are an important in our daily life as they offer us the impression of fruits, foods, fabric and even medicines; and influence people's choice. Colorant can be categorized into three types which are natural, synthetic and inorganic colours. The natural colours are naturally provided by living organisms such as plants, animals and microorganisms.

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Synthetic colours are produced in laboratories and cannot be found in nature. The organic compounds used for this type of colorant can be obtained by chemical synthesis. Inorganic colours are refers to minerals such as gold, silver and titanium dioxide; and can be found in nature or obtained by synthesis [1]. Low price and easy synthesis are among the factors of artificial colorants application in industry. However, the demands on clean label, healthy life style, strict regulation and advance technology trigger the colouring agent replacement into natural sources which caused the utilization of these sources to greatly increase [2]. According to Singh and Tyagi, consumers are willing to pay high price for natural based products due to healthfulness issues [3]. The global market for natural colorant in food industry alone is expected to reach USD 1.7 billion in 2020.

Synthetic colorants tends to be harmful as they can render allergenic and intolerance reactions to human beings [4]. Meanwhile, the colours from natural sources are proved to be safe due to their non-carcinogenic, non-toxic and biodegradable in nature properties [5]. Plant is one of the prime sources of natural colorant and there are lots of reported work and ongoing researches on this matter. Plants have been also used as colouring agent for ages in various industries due to its easily available source in nature and significantly provide health benefits to the consumers [6 - 8]. There are several active constituents in plants which serve as different colour pigments such as anthocyanins, carotenoids, betalains, crocins, anthraquinone and more [9 - 11]. In this review, a thorough summary on the historical background, classifications, natural pigment from plants, processing technologies and its applications in wellness industries are discussed.

History: The historical background of dyeing started since early civilizations where it was practiced since Bronze Age in Europe. The earliest written record for natural colorant dated 2600 BC was found in China [12]. The usage of natural colorant as a dyestuff for textile has been witnessed in the Indus Valley period (2500 BC) by findings coloured garments and traces of madder dyes in the ruins of 3500 BC Mohenjodaro and Harappa civilization. The evidence of colorant in Eqypt was found in the tomb of King Tutankhamen where the red fabrics of wrapped cloth showed the presence of alizarin, a pigment from madder. In Central and North America, the cochineal dye was used by the people of Aztec and Maya culture period. By the 4th century AD, dyes including woad, madder, weld, Brazilwood, indigo and a dark reddish-reddish purple were known [13].

Pigment from henna was used even before 2500 BC, while saffron is stated in the Bible [14]. Use of natural colorants in food is known in the shosoin text of the Nara Period of Japan (8th century) which contains references on soybean and adzuki-bean cakes colouring, thus it provide information that the coloured processed foods had been applied at least by the people of the some sections during that period. In addition to that, the development of natural colours from microorganisms was reported to be started with the production of red rice wine, red shaohsing wine and red Chinese rice using mold (Monascussp.) during 1884. Later, fungi (1954), yeast (1963 to early 1970s) and microalgae (Late 1970s, early 1980s) were being utilized to obtain the colours [15].

The natural colorants have been applied for centuries using primitive dyeing techniques such as sticking plants to fabrics and rubbing crushed pigments into cloth, the methods were emerging with civilizations [16]. However, the introduction of first synthetic dyes from coal tar distillate by Perkin in 1856 trigger the growth of convenient and cheap synthetic pigments to replace the use of natural dyes [17]. According to Lomax and Learner [18], naturally occurring pigments has been completely displaced by synthetic dyes in 20th century such as phthalocyanines, arylides and quinacridones. Approximately 200 synthetic colorants were listed in Food and Drug Administration (FDA) from 1938 to 1960. The demonstration against synthetic colorants usage especially on food products by activists from 1960 to 1970 decreases the number dramatically. As of today, only seven synthetic pigments are approved by FDA regulations which can be used in food pigmentation [19, 20]. Nowadays, natural colorants are gaining attention worldwide and it is expected that the trend of natural colorants will continue in the future. And it is expected that the trend of natural colorants will continue in the future.

Classification of Natural Colorants: Natural colorants are often classified based on their sources, colours and chemical structures of chromophore. First, the natural colours are naturally provided by living organisms such as plants, animals, insects and microorganisms. Plants and microorganisms are considered suitable for biotechnological production of colorants due to their understanding on cultural techniques and processing. Currently, 16 of 40 colorants approved as food additives in EU are from plant origin. Meanwhile in US, 26 natural colorants are legalized to be used without certification [21]. The natural pigments derived from bacteria are expanding nowadays as they reported to have good biodegradability and compatibility with the environment [20]. The pigments from natural sources can also be classified by their colour [22]. Some of important basic colours are blue, red and yellow. According to Gulrajani[14], indigo is the only viable colour for blue. Natural indigo usually obtained by fermenting the leaves of *Indigofera* species and woad. **Red** and yellow are most common colour in natural dyes. Madder and annatto are the example of red colour source meanwhile kamala, turmeric and marigold flower producing yellow pigment. There are many other available colours obtained from plants such as green, white and brown. Basically, the E number used by EU for food colouring was assigned based on their colours.

Third, the natural pigments can be divided into eight classes of the chemical structures basis as in **Table 1**.

TABLE 1: CLASSES OF CHEMICAL STRUCTURES BASIS FOR NATURAL PIGMENTS [23 - 25]

S. no.	Name	Properties
1	Indigoid dyes	Basic structure is indigotin and it is the source of primary colour blue
2	Pyridine based dyes	An isoquinoline alkaloid with a bright yellow colour. Example is barberine
3	Carotenoid dyes	Polymer of eight isopropene monomers which give the colour of yellow, orange and red such as bixin, lycopene and carotene
4	Quinonoid dyes	Quinine functional groups are found in the structure. The colour ranging from yellow to red. Examples are benzoquinones, naphthoquinones, antroquinones
5	Flavonoid dyes	A hydroxyl or methoxy substituted flavone exist in structure. Offers several groups of pigment colour including pale yellow (isoflavones), deep yellow (flavones, aurones), orange (aurones), red and blue (anthocyanins)
6	Dihydropyran based dyes	The structure is closely related to flavones. Main colouring substances are brazilin and haematoxylin
7	Betalain dyes	Consists of water soluble nitrogen in the structure which comprise of betaxanthins (yellow) and betacyanins (violet)
8	Tannin based dyes	The dye has polyphenolic structure and yielding brown in colour

Demands for Natural Colorants: The usages of synthetic colorants in various industries are severely criticized over the years due to health factors, environmental consciousness and consumer awareness [26]. Ahmad *et al*., reported in their article that about 10% of industrial dyes are discharged into the environment and cause harmful effect on environment and living organisms due to high toxicity level [27]. The demonstration in US on synthetic additives in 1960 brought the colorants from nature to light. The pharmacological properties, nutritional characteristics, hygiene and good quality are among the reasons of high interest of natural colorants. However, natural colorants are sensitive towards pH, heat and light [28] which cause them to have lack of stability. To address this issue, microencapsulation and nano-encapsulation of natural colorants have been proposed [11, 29 - 31] and the studies proved that this method improve the crocin and anthocyanins stability under various environmental conditions. In addition, the formulation of natural pigments with other components can also sort out low stability problems. Application of emulsion system in cosmetics is one of the examples [32].

Natural Colorant from Plants: As of 1996, it was revealed that only 0.5% from 70% plants with colours worldwide has been extensively studied. The number is increasing over the years as the interest for natural colorants are growing. This section will discuss some major and common natural colorant agents from plants used for industrial purposes. *Bixa orellana* or commonly known as annatto is one of the oldest natural occurring dye yielding plants. The resinous outer covering of the annatto seed provide reddish orange colour and composed of carotenoid pigments; bixin, norbixin and their esters **Fig. 1** [33]. Bixin accounts more than 80% of total annatto pigments [34]. Annatto presently used in food and textile industries. This plant pigments can also be applied in pharmaceutical field as they have potential in anticarcinogenic activity [35], anticlastogenic activity [36] and exhibit chemo-preventive [37].

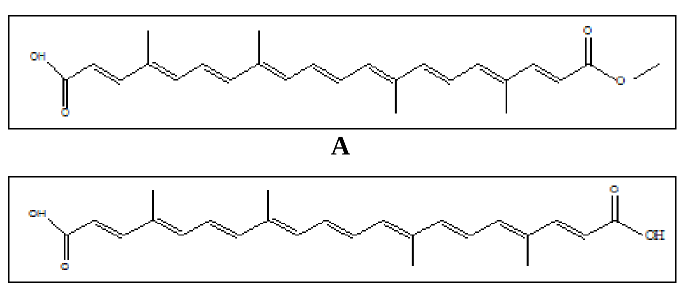


FIG. 1: CHEMICAL STRUCTURES OF (A) BIXIN; (B) NORBIXIN

Turmeric (*Curcuma longa* Linn.) contains 50 to 60% curcumin, which reported to be responsible for yellow colour dye [38]. The unique chemical structure of curcumin as in **Fig. 2** contributes to pharmacological activities of turmeric such as anti-inflammatory, antiviral, antibacterial and powerful antiseptic agent [39].

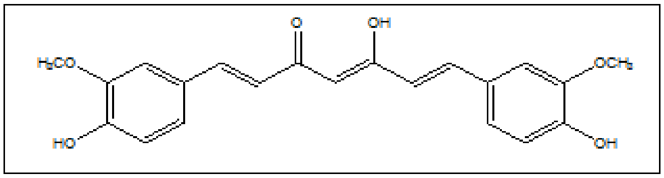


FIG. 2: CHEMICAL STRUCTURE OF CURCUMIN

Lawsonia inermis L. (henna) is widely used in cosmetics industry as dyeing agent and for its medicinal purposes. Henna bound hundreds of phytoconstituents and the prevalent in the extracts are coumarins, flavonoids and naphthoquinones [40]. According to Singh *et al*., as the name suggested, the key colouring matter is 2-hydroxy-1,4-naphthoquinone or commonly known as lawsone (0.4 to 1.5%) which yielding red-orange pigment of henna leaves extract **Fig. 3** [41]. Henna is recognized as medicinal plant because of their antibacterial, antifungal, wound healing and hypotensive abilities [42].

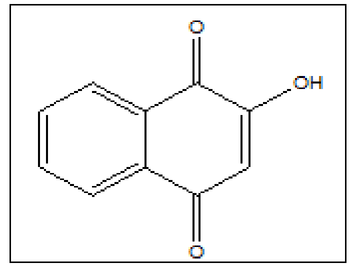


FIG. 3: CHEMICAL STRUCTURE OF LAWSONE

Saffron or *Crocus sativus* Linn. made up from petal, anther and stigma parts with the latter is the important part. Yellow-orange colorant of saffron stigma was contributed by the existence of crocin along with other major constituents namely crocetin, picrocrocin and safranal [33, 39]. The chemical structures of these constituents are illustrated in **Fig. 4**. The therapeutic values of saffron has been studied for years and proven that they exhibit antidepressant, antitumor, antioxidant and have the ability to improve memory [43 - 47]. These advantages promote the use of saffron in food, cosmetics, textile and pharmaceutical industries [10].

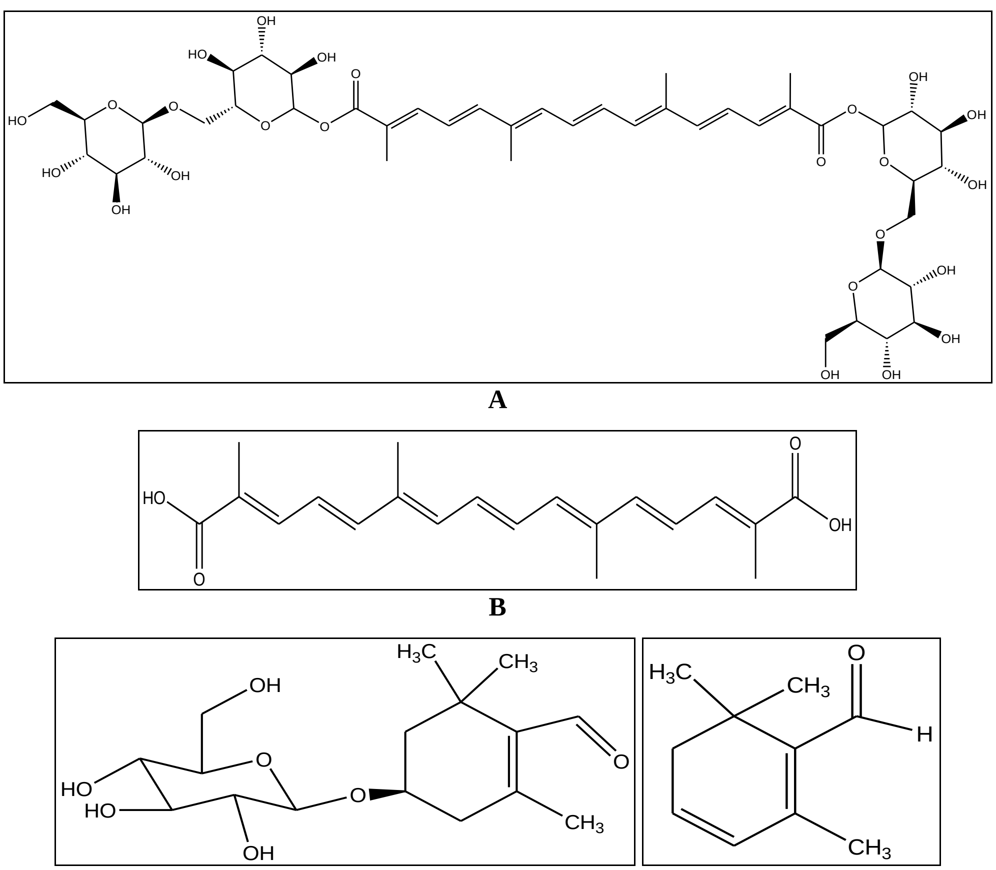


FIG. 4: CHEMICAL STRUCTURES OF (A) CROCIN; (B) CROCETIN; (C) PICROCROCIN; (D) SAFRANAL

Marigold (*Tagetes erecta* L.) is one of the richest natural sources of carotenoids, marigold flower are often extracted for their yellow-orange pigment; *i.e.* lutein **Fig. 5** [48]. The yellow-orange pigment is used as additive to animal feed in order to provide colour to egg yolks. Marigold also widely applied in phytotheraphy, dermatology and cosmetology areas for its inflammatory, antioxidant, analgesic and anti-edematous properties [49 - 51].

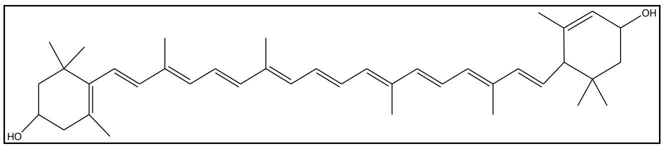
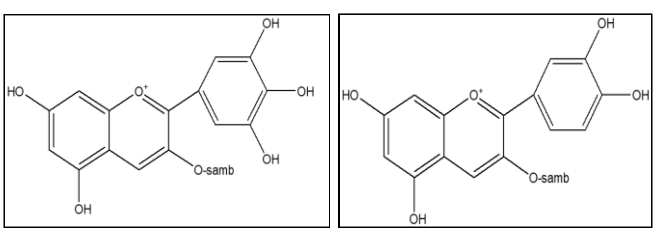


FIG. 5: CHEMICAL STRUCTURE OF LUTEIN

Roselle (*Hibiscus subdariffa*) calyces are rich with anthocyanins, mainly delphinidin-3-sambubioside and cyanidin-3-sambubioside which produced red pigment **Fig. 6** [52]. The colorant from roselle is highly used traditionally as they offer several health benefits including strengthen the bone and teeth formation due to the presence of mineral contents in the extract such as potassium, magnesium and calcium [53]. Besides, it is known that the calyces can treat high blood pressure [54], possess anticancer [55], antibacterial [56] and antioxidant [57] properties. To date, there are several food products which applied red pigment from roselle, for example emulsifier for non-alcoholic drink and juice [58].



A **B**

FIG. 6: CHEMICAL STRUCTURES OF ANTHOCYANINS (A) DELPHINIDIN-3-SAMBUBIOSIDE; (B) CYANIDIN-3-SAMBUBIOSIDE IN ROSELLE CALYCES

Purple carrots (*Daucus carota* L.) were extracted to obtain anthocyanins which contribute to red colour pigment. The natural pigment is often used in food industry as colorant in preparing hard candy and sweet jelly. The antioxidant ability of the pigment is utilized as additive in sunflower oil in order to delay its rancidity [59].

To sum up, most of natural colouring plants showed pharmacological properties which can be applied not only as food colorants, but also in various industries so that the dependency on synthetic colorants can be minimized.

Production Technologies of Natural Colorant: Production technologies of natural colorants from plants could vary from simple solvent extraction to sophisticated techniques depends on the product requirement and its purity. The extraction procedure such as soxhlet method to obtained colour from *Quercus robur* L. [60], supercritical fluid extraction and pressurized liquid extraction on annatto seeds and jabuticaba skins [61], and microwave assisted extraction used on milkweed leaves to get its colour [62] are among several techniques commonly applied. After the extraction process, purification and drying are essential to ensure the quality of the finished product. Purification steps involve filtration, reverse osmosis or preparatory high performance liquid chromatography will be used to eliminate contaminants and increase the purity of the extracts [14]. The common practices to preserve the stabilization of colours are through spray drying or freeze drying [63]. In addition, these procedures is convenience for storage purpose. Further analysis and characterization are other fundamental element need to be done in term of their colour and chemical analysis to evaluate the features of natural colorants.

Implications in Wellness Industries: The development of natural pigments application in wellness industries especially food, cosmetics and pharmaceutical has been established attributed by their nature abilities in various aspects.

Food Industry: The necessity of the use colorants in food substances has been highlighted by Food and Nutrition Board to (1) maintain original food appearance, (2) assure colour uniformity, (3) intensify food's normal colour, (4) protect characteristics of food such as flavour and Vitamins, and (5) increase food acceptability [64]. Colorants from natural sources as food additive are able to prevent food spoilage caused by fungi, viruses and bacteria since most of them, including anthocyanins and carotene, possess antagonistic activity. Carotenoids also can protect the food from intense light hence maintaining the quality of food [26]. The examples of natural colorants in foods are annatto pigment in cheddar cheese [65] and marigold flower extract in poultry products [66].

Cosmetic Industry: Natural dyes are commonly applied in cosmetic industry due to their privileges of no side effects, compatible with all skin types, UV protection, not tested on animals and anti-aging properties [67]. There are numerous cosmetics in skincare, hair care, fragrance *etc*. has been developed. To name a few, yellow pigment of turmeric is a powerful antiseptic for skin revitalization [39] and antioxidant ability of carotenoid from annatto oil provide benefits as body care products besides give sunny colour to creams, shampoos and lotions [7]. Roselle colour is often used as natural colorant in lipstick due to the effectiveness of anthocyanin content in minimizing wrinkles, fine lines and dryness of lips [68]. The orange colour from ß-carotene of carrot oil can metabolize into Vitamin A which plays an important role in anti-aging, revitalizing and rejuvenating agent [69].

Pharmaceutical Industry: Generally in pharmaceutical industry, the formulation of tablets, tablets coatings, capsules, liquid orals, toothpaste and ointment are coloured to increase the aesthetic appearance, prolong the stability, produce standard preparations or for identification purposes [1]. The active constituents in natural pigments are utilized to meet the demand in medicine. For examples, carotenoids are used as Vitamin supplements [70], bixin is applied as one of the natural colouring agents for ointments and plasters and film coating for tablets employed chlorophyll, carotenoids and anthocyanins as basic ingredients.

CONCLUSION: The great attention from the public urges the industries for the continuous search and production of natural colours in replacing the synthetic one. Non-toxic properties and less side effect of natural colorant from plants showed promising results as a source of colouring agent not only because of the colour, but also having wide range of medicinal properties give added values to the natural pigment. Despite the demand growth and consumer awareness towards natural ingredients in consumable products, handling and stability issues are still a hindrance. By knowing the limitations of each natural pigment, appropriate precaution steps can be considered. Incorporation of modern technologies to cope with drawbacks and safety concerns of natural colorants is needed for further development in wellness industry so that those abundance natural sources are not wasted. More detailed studies are needed on availability, characterization and documentation of colour yielding plants to evaluate their real potential for daily usage and commercialization purposes.

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