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REVIEW OF ECO FRIENDLY AND PROTECTIVE NATURAL DYES

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ABSTRACT

Current review addresses eco friendly and protective natural dyes. Sources of natural dyes and their extraction from plants, insects and microorganisms are described. Development of natural dyes within the scope of their application in textiles was studied In this regard, mention is made of mordanting, dyes for healthy eco friendly and sun protective textiles. Antimicrobial activity of some natural dyes along with their medicinal effects are also mentioned. Natural dyes are derived from naturally occurring sources such as plants (e.g., indigo, curcumin and saffron); insects (e.g., cochineal beetles and lac scale insects). A spectrum of beautiful natural colors ranging from yellow to black exists in the above sources. New strategy for development of non toxic antimicrobial textiles is additionally outlined. Fastness properties such as light fastness, Washing, rubbing fastness are studied for textile fibers.

KEY WORDS: Natural dyes, sources, extraction, textile fabrics, antimicrobial activity.

INTRODUCTION

Textile processing industry is one of the major environmental polluters. The effluent generated by much water used would pollute the environment as it contains a heavy load of chemicals including dyes used during textile processing. There are two main methods to limit the environmental pollution of textile processing. One is to construct sufficiently large and highly effective effluent treatment, and the other method is to make use of natural dyes and chemicals that are environment friendly.[1,2]

A renewed international interest has focused on natural dyes due to increased awareness of the environmental and health hazards associated with the use of synthetic dyes. Natural dyes are derived from naturally occurring sources such as plants (e.g., indigo, curcumin and saffron); insects (e.g., cochineal beetles and lac scale insects). A spectrum of beautiful natural colors ranging from yellow to black exists in the above sources. These colors are exhibited by pigments and their mixtures are due to the absorption of light in the visible region of 400-800 nm. This absorption of light depends on the structure or constituents of the coloring pigment/ molecules contain various chromospheres present in the dye display the colors. Natural dyes are mostly eco-friendly, biodegradable, less toxic, and less allergenic as compared to synthetic dyes. Most of the natural dyes are safe and some even have curative effect e.g., curcumin in turmeric has antibacterial properties[3] Several synthetic dyes have been banned because they cause allergy-like symptoms or are carcinogens. Nowadays, natural dyes are commonly used in the cosmetic industry due to no side effects, UV protection and anti-aging properties.[4]

Natural dyes can be divided into two classes such as substantive and adjective (non-substantive). Substantive dyes are chemically bounded to the fiber without the aid of any color fixing agents (mordants) as., indigo, turmeric. Most of the natural dyes being non-substantive are used with mordants.[5]

Mordant forms a complex between fiber and dye, which is insoluble in water and thus gives a fast color. Some of the adjective natural dyes are madder, logwood, catechu, lac, cochineal, kermes, beetroot and marigold, rose.[6,7]

Metal ions of mordants act as electron acceptors for electron donors to form co-ordination bonds with the dye molecule, making them insoluble in water.[8] Alum, chrome, stannous chloride, copper sulphate, ferrous sulphate etc. are the commonly used mordants.[9,10,11]

Many natural dyes are considered as antimicrobial agents owing to the presence of a large amount of tannins. Several other sources of plant dyes rich in naphthoquinones such as lawsone from *Lawsonia inermis* L.(henna), and lapachol from alkanet are reported to exhibit antibacterial and antifungal activity.[11,12] Some natural dyes have medicinal properties. Some of its contacts are anti-allergic and proved to be safe for body contact. Majority of natural dyes are non-toxic. Natural dyes find use in the colorations of textiles, foods, drugs, and cosmetics. Small quantities of dyes are also used in coloration of paper, leather, shoe polish, wood, cane, candles, etc. In the earlier days, dyes were derived only from natural sources.

1. Sources, Types and Characterization of Natural dyes

A dye can be defined as a highly colored substance used to impart color to variety of materials like textiles, paper, wood, varnishes, leather, ink, fur, foodstuff, cosmetics, medicine, toothpaste, etc. As far as the chemistry of dyes is concerned, a dye molecule has two principal chemical groups, as chromophores and auxochromes. The chromophore, usually an aromatic ring, is associated with the coloring property. It has unsaturated bonds known as auxochrome such as –C=C, =C=O, –C–S, =C–NH, –CH=N–, –N=N– and –N=O, whose number decides the intensity of the color. The auxochrome helps the dye molecule to combine with the substrate to produce color.[13,14]

Natural dyes comprise those colors that are obtained from animal or vegetable matter without chemical processing. Natural dyes fall into the following categories: 1-Leaves and stems 2- Twigs and pruning 3- Flower heads 4- Barks 5- Roots 6- Outer skins, 7-hulls and husks 8- Heartwoods and wood shavings 9- Berries and seeds 10- Lichens Insect dyes. Many plants and some animals' are potentially rich in natural dye content. Various parts of plants like roots, stems, bark, leaves, fruits and seed contains coloring matter.

2. Classification of Natural dyes

Natural dyes are classified based on their chemical structure, sources (Table 1), method of application, color, etc. Based on their chemistry.[17] They are classified into the following groups based on chemical structure

Indigo dyes

This is considered to be the most important dye obtained from the plant *I. tinctoria* L.

Anthroquinone dyes

Some of the most important red dyes are based on the anthroquinone structure. These are obtained from both plants and insects. These dyes have good fastness to light. They form complexes with metal salts and the resultant metal–complex dyes have good fastness.

Alpha-hydroxy naphthoquinones

The most prominent member of this class of dye is henna or lawsone (*L. inermis* L.).

Flavones

Most of the natural yellow colours are hydroxyl and methoxy derivatives of flavones and iso flavones.

Dihydropyrans

Closely related to flavones in chemical structure are substituted dihydropyrans.

2.1. Natural dyes from plants

Many natural dyes were obtained mainly from plants. Almost all parts of the plants like root, bark, leaf, fruit, wood, seed, flower, etc. produce dyes.[18,19] Sources of different colored dyes and mordants are illustrated in (Table 1).

Table 1: Sources of different colored dyes and mordants.

Colour	Botanical name	Parts used	Mordants
Red dye			
Safflower	*Carthamus tinctorius* L.	Flower	–
Caesalpinia	*Caesalpinia sappan* L.	Wood	Alum
Madder	*Rubia tinctorium* L.	Wood	Alum
Log wood	*Haematoxylon campechianum* L.	Wood	–
Khat palak	*Rumex dentatus* L.	Wood	Alum
Indian mulberry	*Morinda tinctoria* L.	Wood	Alum
Kamala	*Mallotus philippinensis* Muell.	Flower	Alum
Lac	*Coccus lacca* Kerr.	Insect	Stannic chloride
Yellow dye			
Golden rod	*Solidago grandis* DC.	Flower	Alum
Teak	*Tectona grandis* L.f.	Leaf	Alum
Marigold	*Tagetes* sp.	Flower	Chrome
Saffron	*Crocus sativus* L.	Flower	Alum
Flame of the forest	*Butea monosperma* (Lam) Taubert.	Flower	Alum
Blue dye			
Indigo	*Indigofera tinctoria* L.	Leaf	Alum
Woad	*Isatis tinctoria* L.	Leaf	–
Sunt berry	*Acacia nilotica* (L.) Del.	Seed pod	–
Pivet	*Ligustrum vulgare* L.	Fruit	Alum and iron
Water lily	*Nymphaea alba* L.	Rhizome	Iron and acid
Black dye			
Alder	*Alnus glutinosa* (L.) Gaertn.	Bark	Ferrous sulphate
Rofblamala	*Loranthus pentapetalus* Roxb.	Leaf	Ferrous sulphate
Custard apple	*Anona reticulata* L.	Fruit	–
Harda	*Terminalia chebula* Retz.	Fruit	Ferrous sulphate
Orange dye			
Annota	*Bixa orellena* L.	Seed	Alum
Dhalia	*Dhalia* sp.	Flower	Alum
Lily	*Convallaria majalis* L.	Leaf	Ferrous sulphate
Nettles	*Urtica dioica* L.	Leaf	Alum

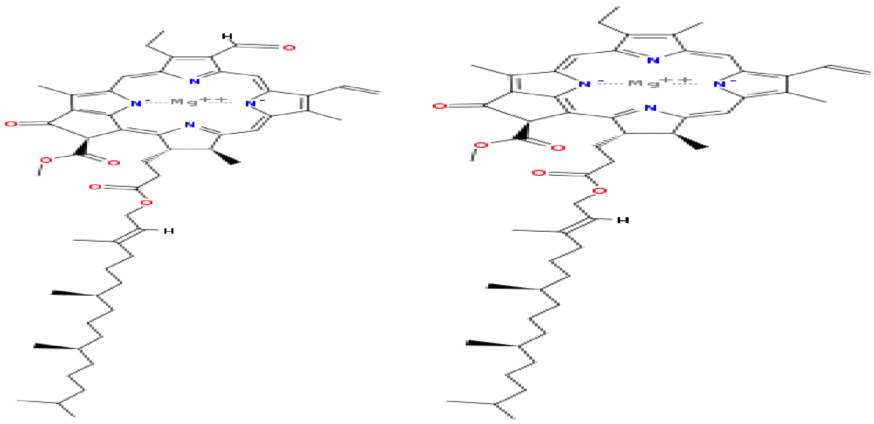
2.2. Natural dyes from microorganisms

The production and evaluation of microbial pigments as textile colorants is currently has been investigated**.** Fungi are more ecological interesting source of pigments, since some fungal species are rich in stable colorants such as anthraquinone. Anthraquinone derivatives were previously isolated from the fungus *Dermocybo Sanguinea* .A number of anthraquinone derivatives have been identified from various species of fungi and Lichens. These metabolites are of interest because many of them possess significant antibiotic activity, primarily ainst gram- positive bacteria and *Pseudomans aruginose.* Due to the rapid population growth, industrialization & urbanization, there are more than 10,000 different synthetic dyes widely used in the textile, paper, cosmetics, food and pharmaceutical industries.

It is advantageous to produce pigments from microorganisms rather than plants, due to their fast growth in a cheap culture medium, independence of weather conditions and colors of different shades].

3. Extraction of some natural dyes

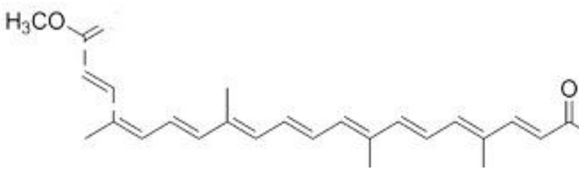
a) chlorophyll can be extracted from leaves and green stems of plants using acetone in presence of $CaCO_3$ or any mild alkali. The weak bases neutralize any acid, that may be liberated from the tissue and prevents the production of pheophytins during extraction process. Usually these extracts can be assayed at 654 nm using UV/visible spectrophotometer. chlorophyll was a mixture of two compounds, chlorophyll-*a* and chlorophyll-b[20] studied the effects of ultrasonic energy on dyeing silk fabrics with Chlorophyll natural dye using ultrasonic method. The results of investigation offer a new viable method for dyeing of silk fabrics by environmentally friendly method and saving energy.[21]



Structure of Chlorophyll(b) Structure of Chlorophyll (a)

Figure 1: Chemical Structure of Chlorophyll (b) and Chlorophyll (a).

b) Carotenoid pigmets (yellow to red colour) are exracted from carrot, tomato and antto usually by acetone: Hexane (1:3) mixture. Acetone layer is removed by washing the extract continuously with water. The hexane extract is then treated with activated MgO_2 diatomaceous earth column, eluted with acetone and hexane. The acetone fraction usually contain polar xanthophylls and the hexane eluate contain non polar carotenoids. The carotenoid can be detected using a UV-spectrophotometer with a lmax value ranges from 430nm to 480nm (eg: b-carotene l max is 453nm in hexane).



Chemical structure of carotenoid pigment

c) Beatline constitute another group of colour pigments from plants. Betalains are present in two forms, one is b-cyanin which is purple red and found in high concentrations and the other one is b-xanthine which is yellow in colour and found only in low concentrations. Betalains are highly water soluble therefore it can be extracted easily with water from plant tissues. The water extract is then blended in ethyl alcohol : water (1:1) mixture. Ethanol in the mixture reduce the enzymatic action which is otherwise cause degradation of pigments. The extract can be screened for the presence of betanin and vulgaxanthin by the absorption maximum of there compounds at 535-540nm and 476-478 nm.

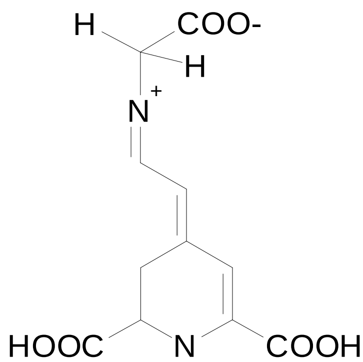


Figure 2: Chemical structure of Beatline pigment.

d) Anthocyanins: These colour pigments are present abundantly in plant kingdom, responsible for imparting purple, red and blue colour, of flowers, fruits and vegetables Anthocyanins used to show specific colours at a particular pH. These plant pigments are soluble in water and can be extracted easily from plant parts, by using slightly acidified water (0.05% HCl). Acidity in water prevent hydrolysis of anthocyanins in the extract. Usually extract containing anthocyanins with pH 1.0 show absorption maximum at wavelength 510 to 540 nm. Red cabbage contains pigments called anthocyanins. These pigments give the purplish colour. On the right side is the general chemical structure of anthocyanins. R1 and R2 can be different chains, like H, OH and OMe.

	R_1	R_2
Cyaniding	H	OH
Delphinidin	OH	OH
Malvidin	O-CH3	O-CH3
pelargonidin	H	H
Peonidin	H	O-CH3
Petunidin	OH	O-CH3

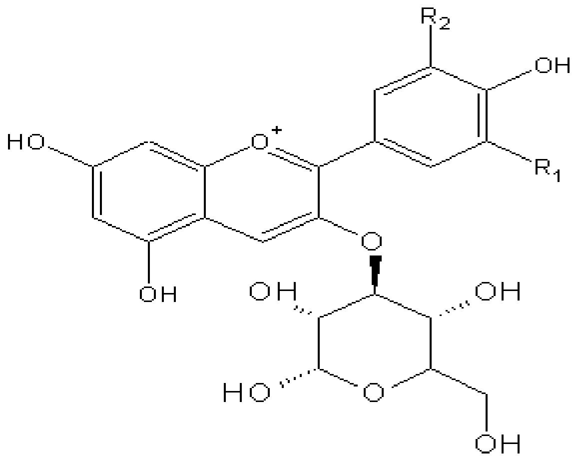


Figure 3: Chemical structure of Anthocyanins pigment.

3.1. Ultrasound for Extraction of Natural Dyes

Ultrasound is classified according to frequency range as power ultrasound (20–100 kHz) and diagnostic ultrasound (1–10 MHz). When a liquid is irradiated by ultrasound, micro bubbles appear, grow and oscillate extremely quickly and even collapse violently if the acoustic pressure is high enough. The occurrence of these collapses near a solid surface will generate micro jets and shock waves. Moreover, in the liquid phase surrounding the particles, high micro mixing will increase the heat and mass transfer and even the diffusion of species inside the pores of the solid.[22]

Procedures of extraction

1 g of the plant samples was taken and added to 50 ml water taken in the beaker. The beaker was covered using aluminum foil to prevent loss of solvent by evaporation. The ultrasonic probe was positioned in the beaker and the ultrasound power set at 80 W. Since the ultrasonic bath temperature raise from room temperature 30 ◦C to 50◦C in 3 h time, the temperature was maintained at around 45 ◦C by using a water bath without any external heating. Samples were taken at every 30 min and the optical density was determined with the help of UV–VIS spectrophotometer. At the end of 3 h, the yield and extraction efficiency of each sample was determined by gravimetric method. The filtrate was tightly closed and stored at low temperature.[22] In the case of natural dye extraction, ultrasound is used as a tool for enhancing mass transfer of coloring matter from natural plant material and transport to the solvent medium., ultrasound technique could be beneficial for extraction of natural dyes and subsequently for leather dyeing and it is also used in textile dyeing.[22,23]

Mordanting

Few natural dyes are color-fast with fibers. Mordants are substances which are used to fix a dye to the fibers. They also improve the take-up quality of the fabric and help improve color and light-fastness, indigo for example, will fix without the aid of a mordant; these dyes are known as 'substantive dyes'. Others dyes, such as madder and weld, have a limited fastness and the color will fade with washing and exposure to light.[10] Mordants used such as alum, copper sulphate, iron or chrome (there are concerns, however about the toxic nature of chrome and some practitioners recommend that it is not used).

4. Development and application of Natural Dyes on Textiles

The use of natural dyes faded drastically for the last four to five decades and dyeing practice with the synthetic compounds enhanced many folds, without thinking much about the dreadful and silent killing effects of the synthetic colors on the environment and eco systems. The production and processing of the synthetic dyes and colorants were estimated to release many hundred tones of unfixed and non degradable wastes, which are hazardous to human health, also might cause major environmental pollution and ecological imbalance. Perhaps this may be the fact for the new awareness and resurgence of interest all over the world towards natural resources for getting biodegradable and environmental friendly dyes. After dyeing the color of the fabric should not be affected during washing process, dry cleaning with organic solvents etc and also the dye should give fastness to light, heat and bleaching.[10] Natural dyes, are usually applied to color natural fibers, Natural fibers come mainly from two distinct origins, animal origin or vegetable origin. Fibers from an animal origin include wool, silk, mohair and alpaca, as well as some others which are less well known. All animal fibers are based on proteins. Natural dyes have a strong affinity to fibers of animal origin, especially wool, silk and mohair and the results with these fibers are usually good. Fibers of plant origin include cotton, flax or linen, ramie, jute, hemp and many others. Plant fibers have cellulose as their basic ingredient.

4.1. Eco-friendly sun-protective clothing

Although most people view sunscreen as their first line of defense against the sun, what you wear can really play a major role in safeguarding your skin. It is found that donning sun-protective clothes and scaling back sun exposure beats out sunscreen as a skin cancer prevention strategy.[24] And, if you choose eco-friendly garments—those that are certified organic, made from renewable materials, and unbleached—you can double your green impact: keep petrochemical-based sunscreens from entering the environment.[24] There are some rules for choosing clothes that will protect skin from the sun as:

Eco-friendly sun-protective clothing helps you go green because wearing sun-protective clothing instead of slathering on the sunscreen will keep health- and environment-endangering petrochemicals out of the environment.

Organic cotton farming keeps thousands of pounds of toxic pesticides and insecticides out of the environment each year. Undyed clothing, or that colored with plant-based dyes, keeps chlorine and other toxic bleaching agents.

4.2. Antimicrobial activity of some natural dyes

Textile materials and clothing are known to be susceptible to microbial attack, as these provide large surface area and absorb moisture required for microbial growth.[23] Natural fibres have protein (keratin) and cellulose, etc., which provide basic requirements such as moisture, oxygen, nutrients and temperature for bacterial growth and multiplication. This often leads to objectionable odour, dermal infection, product deterioration, allergic responses and other related diseases**.** This necessitates the development of clothing that could provide a desired antimicrobial effect.

Many of the plants used for dye extraction are classified and some of these have recently been shown to possess remarkable antimicrobial activity.[25] *Punica granatum* and many other common natural dyes are reported as antimicrobial agents owing to the presence of a large amount of tannins. Several other sources of plant dyes rich in naphthoquinones such as lawsone from henna, juglone from walnut and lapachol from alkannet are reported to exhibit antibacterial and antifungal activity.

With the advent of improved human life, a new area has developed in the realm of textile finishing. The control of microorganisms, e.g., bacteria, mildews molds, yeasts and viruses on textile fabrics extends into diverse areas as hospital, environment and every day household. Neither natural nor synthetic fibers have resistance to microorganisms. Thus, various antimicrobial finishes and disinfection techniques have been developed for all types of textiles.[26]

Antimicrobial finishes are applied to textiles for three major reasons: (a) to control the spread of disease and the danger of infection following injury, (b) to control the development of odour from aspiration, stains and other soil on textile materials, and (c) to control the deterioration of textiles, particularly fabrics made of natural fibres caused by mildew. In recent years, antimicrobial agents that have been used industrially included quaternary ammonium salts, metal salts solutions and antibiotics. Unfortunately, some of these agents are toxic and poorly effective. This; interpretation, has leads to a special focus on "green chemistry" by researche world-wide.[27]

Green chemistry was strongly created as results of increasing awareness about the environment and welfare.[28] Utilization of nontoxic chemicals environmentally benign solvents and renewable materials are some of the key issues that merit important consideration in a green synthesis strategy.

Chitosan is as an ideal antimicrobial agent.[29] Chitin, the second most abundant biopolymer in nature next to cellulose, is a high molecular weight linear polymer of 2-acetamide-2-deoxy-D-glucopyranose units linked together by 1,4-glucosidic bonds. Chitosan is the deacetylated derivative of chintin It is characterized by non toxicity, biodegradability and compatibility with other ingredients. Previous reports.[12] disclosed that D-glucoseamine hydrochloride (chitosan monomer) dist not slow any growth inhibition against several bacteria whereas chitosan was effective. The antimicrobial activity of chitosan is, therefore, related to not only its cationic nature but also to its chain length. It has been also reported that many plants used for dye extraction are classified as medicinal, and some of these have recently been shown to possess remarkable antimicrobial activity.[30]

CONCLUSION

Natural dyes is used as a substitute of synthetic dyes for dyeing different kinds of textiles to obtain antimicrobial and smart textile.

The use of antimicrobial textiles may significantly reduce the risk of infections especially when they are used in close contact with the skin or in the immediate and non-immediate surroundings.

Dyeing with natural dyes extracted from desert plants, insects and fungi to overcome environmental pollution, natural dyes have antimicrobial activity against various types of microbes as *(Escherichia coli, Staphylococcus aureus and Pseudomons aeruginosa)*, Using nano materials for some for treatment of fabrics before dyeing, to acquire new properties as antimicrobial activities against bacteria and fungi and also to protect textiles from ultra violet rays. Using a traditional and microwave heating for extraction of dyes and dyeing methods because microwave heating is a more effective method than traditional heating. Other additional features about microwaves are that they are cheaper, more economical (saving time and energy), eco-friendly, and produce a higher dye uptake as compared to conventional techniques, Environmentally friendly pre-treatment by chitosan before dyeing in order to obtain dyed fabric with high quality and more protected against microbes. Application of antimicrobial agents in the development in the textiles as chitosan, qutenary ammonium salt and neem.

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