



ANTIBACTERIAL ACTIVITY AND RELATED ISSUES OF NATURAL DYES IN TEXTILES, THEIR EFFECTIVE DATA ANALYSIS

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Abstract: Dyeing textiles with various shades of colors is known to mankind since times immemorial. Indigo and madder root are the most popular natural dyes. Their popularity is relegated after the discovery of azo dyes etc., based on naphthalene anthracene, acridine etc, which were developed as synthetic dyes. Synthetic dyes are hazardous to health and environment and lot of colored effluent is or released into environment. Natural dyes mostly extracted from edible plant sources are safe. A review of some promising plant natural products as dyes, use of metal mordant to fix dyes with textiles and mechanistic approach of mordent dye textile and habitual antibacterial activity are specified in this paper.

Key words; Natural Dyes and Textiles, Mordant and methods, Natural dyes abundant, Antibacterial and dyed textiles

Introduction: In India, as a conservative estimate, no less than 450 plants have dyes called pigments either in leaves, flowers, barks, fruits or rootsⁱ. Many plant pigments are found in edibles like anthocyanines, carotenes, flavanoids glycoside pigments and chlorophyll. They are not suitable as dyes to color textiles, due to their water solubility, rapid photo degradability or lack of functional groups which adhere or absorb to the textile fiber. Natural dyes can be sorted into three categories. Natural dyes (fig-1) obtained from plants (Indigo), those obtained from animals (cochineal), and those obtained from minerals (ocher, all the ochers is iron (III) oxide hydroxide known as limonite)ⁱⁱ. Although some fabrics such as silk and wool can be colored simply by being dipped in the dye, others such as cotton require a mordantⁱⁱⁱ. A mordant is an element which aids the chemical reaction that takes place between the dye and fiber so that the dye is absorbed.

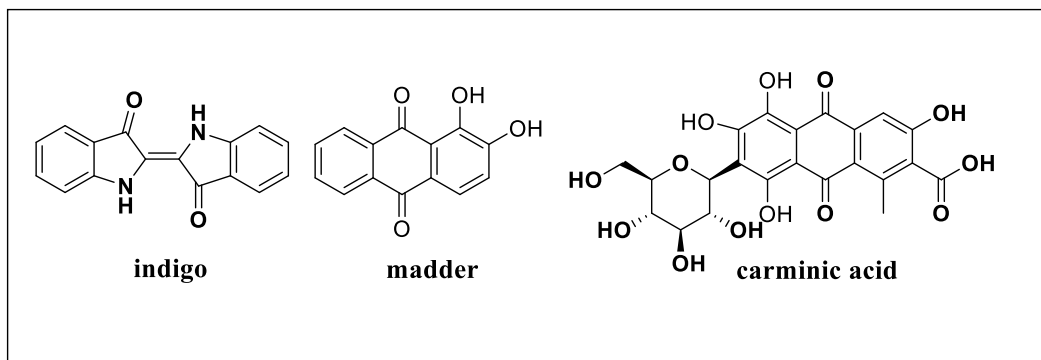


Fig-1

Further even if the dyes functional group adhere to the fiber, mordant is often required as additive. A mordant is a compound which facilitates the chemical reaction between the reactive functional group of dye and the fabric. Common mordants are alums (Ferric ammonium salts) used in conjunction with cream of tartar, which acts as a spread^{iv,v}. The purpose of cream of tartar is to spread the uniformity of color and also to a lesser extent increase brightness. Common mordant are tannic acid, alum, urine, chrome alum, sodium chloride and certain salts of aluminium/iron/copper/tin or chromium^{vi}. However, the most important mordant of choice is alum. The most important colorant in madder (*Rubia tinctorum*) are anthraquinones, Alizarin, Purpuroxanthin, rubiadin, manjistin, purpurin and pseudopurin (fig-2)

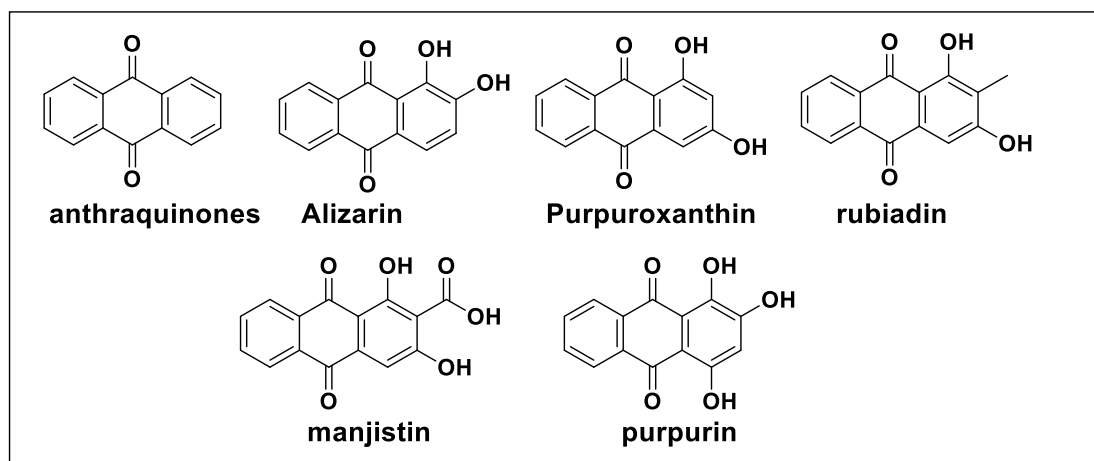


Fig-2

A typical interaction of purpurin a constituent of Manjistin with mordant is given below as described by Ashish Kumar etal^{vii}.

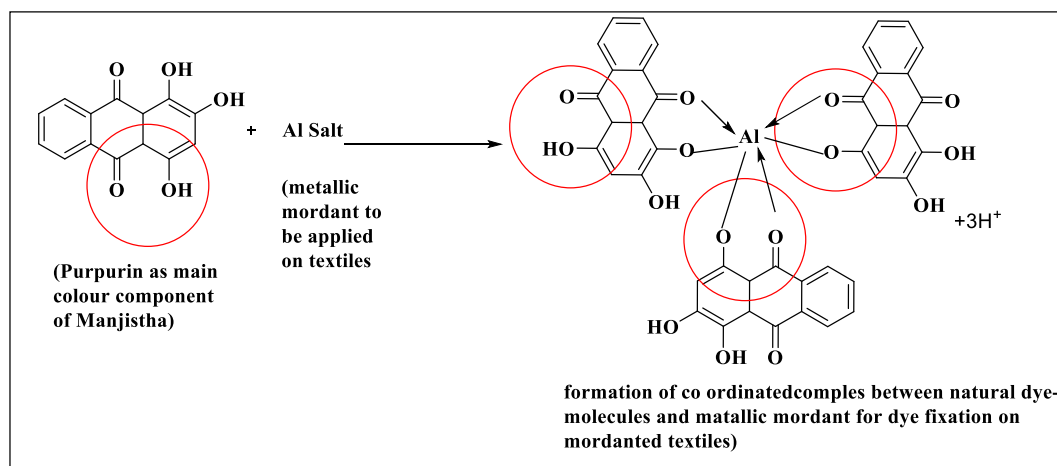


Fig-3

A fifteen step detailed process for dyeing cotton fabric with natural dyes is summarized in a review paper by Ashish Kumar Samanta et al., The cited ref by this author is Mohenty, B.C. *et al.*, Natural dyeing processes of India, pub by calico museum of textiles^{viii}. In this process one of the main steps is pretreatment of cloth with extract of chebulic myrobalan fruit (*Terminalia Chebula*) extract or thani fruit to get yellow strain. This process is called tanning since these fruits contain gallotannins or ellagitannins. This treatment is called galling. After galling, mordanting by potash alum was done and then final dyeing with aqueous extract of a natural dye. Several natural dyes were mentioned in this article.

In "Dye-Mordant-Textile-Fiber" system the salient features of the complex are i) the natural dye cannot be fixed on cellulose without intervention of mordant^{ix}. ii) mordant are metallic salts of aluminum chromium copper etc., iii) the mordant form metal complexes with fibers and the dye mordant (metal salts) first anchor with fabrics and then in turn the anchored metal with fiber and subsequently form a bridge with dye by forming a co-ordination complex^x. iv) While how a dye with functional groups likes C=O (carbonyl) and O-H (hydroxyl group) forms a co-ordination complex is clear as shown in Fig-3.

Types of mordant

Metal salts of aluminum, chromium, Iron, Copper, tin and Potash alum are best choice while chromium salt, potassium dichromate are harmful. Tin salt, stannous chloride gives brighter color but they are prone to oxidation. It is not considered safe from the toxicity point of view. The chemical interaction of bonding to fibers is not clear. It may involve direct bonding hydrogen bonds and hydrophobic interaction. The intermediary mordant form a co-ordination complex with metal complex. This complex involves with C=O (carbonyl) with ortho hydroxyl as shown above in fig-3. Two ortho hydroxyl nitroso (N=O) or azo group of dye form complex with metal ion of mordant. This mordant dye combination in turn may interact chemically with hydroxyl group or hydrogen bonding. However this mechanism is not known but probable structure is given^{xi}. Cited reference in this article^{xii,xiii}

Chemical - Structural features of Dye:

The structural features include extensive conjugation like in carotene which is in turn attached to alum. These are called chromophores shown in circle in chemical structures of chart. The chromospheres group in turn attached to -OH, -NO₂, N=N, N=O, -COOH called auxochromes which enhance the color. Dyes absorb visible light 380 nm to 600 nm, and mostly absorb 550 nm. Pigments are also used in inks, paints and cosmetics.

Now the possible interaction of cellulose hydroxyls with Al-Dye complex may be as shown below. This mordant (Aluminum alum) fix the dye too strongly to textile. The synthetic textile dyes represent a large group of organic compounds that could have undesirable effects on the

environment, and in addition, some of them can pose risk to humans. The increasing complexity and difficulty in treating textile wastes has led to a constant search for new methods that are effective and economically viable. However, up to the present moment, no efficient method capable of removing both the color and the toxic properties of the dyes released into the environment has been found. The color of the textile after fixing Dye-mordant on textile is stable with stand washing and do not fade away. In addition it may exhibit antibacterial activity, if proper selection of the dye is not made. Different dyes impart different color shades. The fixing of shades of the color on textile is a science by itself often by hit and trial. The color depends on the nature of the dye, may be on concentration and the nature of mordant Al, Cr or Sn salts.

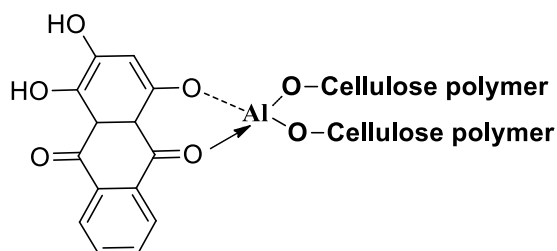


Fig-4 Dye- mordant-Cellulose cotton textile complex

The synthetic dyes both in preparation release colored effluents harmful to environment synthetic dyes may have carcinogenicity. Apart from coloring the fabrics with dye which definitely provide aesthetic sense beside prevent the fading away of white colored fabric. A successful dye should provide antimicrobial properties also. The antimicrobial properties include antibacterial, antifungal properties. It is also desirable that the colored dyed fabric should also resist the insect attack, especially when stored in box. These biological properties of the dyed fabric should be provided by dye itself or by dye and mordent together. Red, blue, green, pink in fact all colors could be obtained by selecting the dye. In addition by changing the concentration of dye and mordant even combination shades could be obtained by selecting the dye. In addition by changing the concentration of dye and mordant even combination shades could be obtained.

Dyed textiles with antimicrobial/antibacterial properties have several uses; a) to protect people against the attack of bacteria and fungi which are present in environment; b) to prevent the spread of microbes from one person to the other. Since times immemorial man used plant pigments for various uses like draw painting to paint houses, utensils, walls and clothes. Natural dyes isolated/extracted from plant sources are safe mostly derived from edible fruits and flowers are non toxic and do not have carcinogenicity.

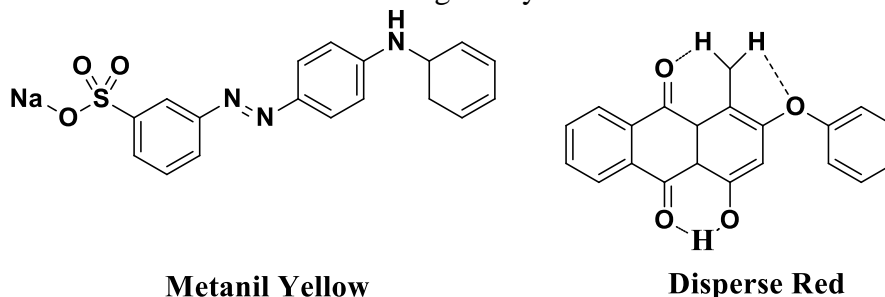
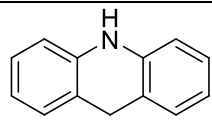
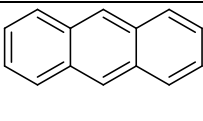
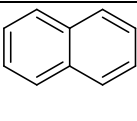
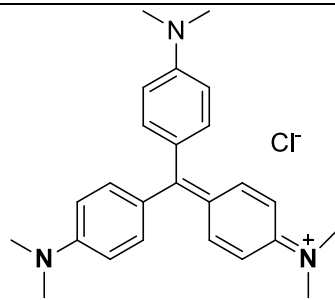
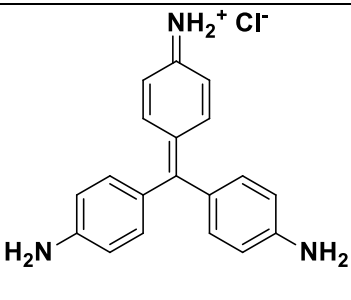
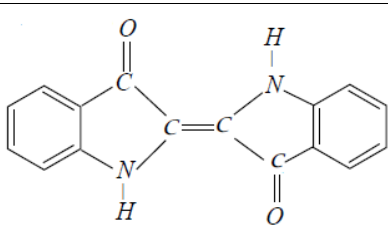


Fig-5

The most important natural dye used commercially is derived from leaves and stems of *Indigofera tinctoria*, by fermentation process. India at one time dominated the export of indigo to British, until the structure and synthesis of indigo elucidated. Until the middle of nineteenth century natural pigments like indigo dominated. The natural pigments relegated after the

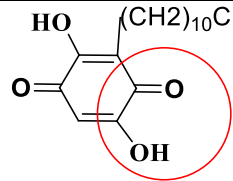
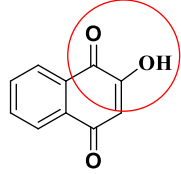
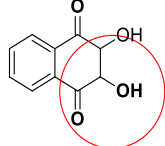
discovery of azo-dyes, anthracene based, naphthalene based and acridine based with sulphonyl group and or azo-based dyes (-N=N-) came into domination. Today the use of synthetic dyes (a typical ref: Synthetic Dyes by Venkataramana, Merck index) are dominating. There is no large scale use of any natural dye. As already pointed out these synthetic dyes are associated with several environmental hazards like releasing toxic effluents into streams, high energy synthetic sequence and often carcinogenic. Table I: gives parent ring system of some typical synthetic dyes (source: Merck index).

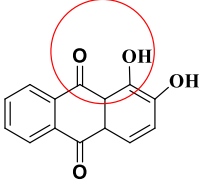
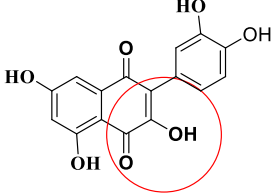
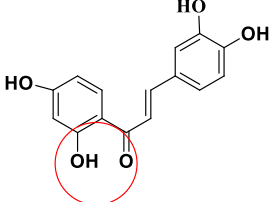
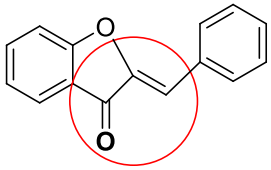
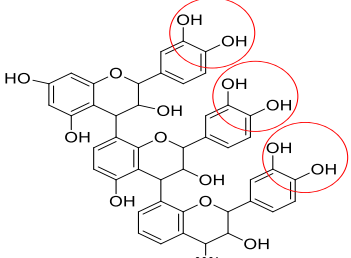
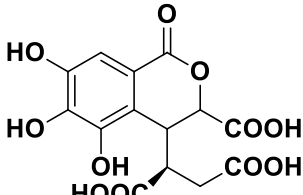
Table-I

		
Acridine	anthracene	naphthalene
		
Crystal violet, antibacterial /dye	Para rosaniline or methylene blue (dye, anti malarial possibly anti bacterial)	Indigo dye

There is normal interest to review natural dyes with antibacterial properties associated with dye textile. The most plant natural dyes belong to the class of natural product called quinones (embelin) benzoquinone (lawsone) anthroquinone (alizarin) flavones (quercetin) chalkones (butein) auronas (meaning gold color). The examples structure portion responsible for dye activity functional group, bound through mordant is shown in circle.

Table-2: structures, names and plan source of some promising natural dyes

	Embelin isolated Berris of <i>Embelicaribes</i> found in Vayavidangka quinone structure
	Lawsone isolated <i>Lawsoniainermis</i> leaves extensively as a coloring agent to decorate the palm of ladies. Benzoquinone structure.
	Plubagin isolated <i>Plumagagozeylanica</i> benzoquinone structure.

	<p>Alizarin <i>Rubia tinctoria</i> most popular dyeing agent for textile fabric</p>
	<p>Quercetin flavonol found in free state or glycoside bound to hydroxyl group. For example rutin a quercetin-3-glycoside found in <i>Fagopyrum esculentum</i> in large quantity (black wheat) 3-OH is quercetin, 3-rutinoside is rutin.</p>
	<p>Butein a chalcone found in flowers of the <i>Butea frondosa</i> flowers. Very common in the forest of Telangana used as holi color. A promising dye for further studies</p>
	<p>Butein, tutin, isobutrin, coreopsin, isocoreopsin aurone also found in <i>Butea frondosa</i> flowers</p>
	<p>Tannins - found in Acacia and other barks used as mordant or dye. Tannins are polymers of catechin units. Tannins act as mordant or dye also.</p>
	<p>Ellagitannins found in myrobalans, arjuna, terminalis, myrobalans promising dyes.</p>

Butein a chalcone found in flowers of *Butea frondosa*, common in the forest of Telangana is used as holi color. All the structures show extensive conjugation number of phenolic hydroxyl shown in circle which is must for dyes. The dyes containing reactive functional groups circled in table-2, chemically bind through chelation or coordination with textile. Most of these classes of compounds are reported to have anti-bacterial activity, for example chalcones. As earlier pointed, the colored compounds like anthocyanin, carotenes, and butaines are water soluble, rapidly photodegradable and are not suitable as dyes.

Based on the above literature and bibliography survey, the following compounds from plants are most suitable for commercial exploitation.

1. Embelin
2. Lawsone
3. Butein
4. Sulfurein (aurone)

5. Tannines (Polymeric molecular weight compounds in *Acacia cassia* and several other plants in bark and heart wood).

6. Ellagitannins-found in myrobalans.

All these plant products are commercially available in large quantities and could be cultivated or propagated. Methods could be established to get extracts from plants containing active constituents dye or pure chemical compound as dye or organic solvent extraction or often simply by water extraction are used to get dyes from the plant products. Different dyes recommended above produce different colors but shades of the color, bright or strong red, brown, blue green, orange red by altering the concentration of dye or mordant or changing the mordant. This could be done by a series of trials.

Many natural pigments are studied form their antimicrobial activity. However, the most desirable or suggested method is to study the antibacterial, antifungal activity of textile fibers treated with the natural dyes. In this connection the paper on antibacterial activity of textiles ,Fabrics treated with Red Pigment from Marine Bacteria by Shiva Krishna etal., is worth mentioning^{xiv}.

There is no literature evidence on the preparation of textiles with the above mentioned selected dyes. In India, during wedding, the couple use yellow tainted dhotis, saris dipped with turmeric water (curcumin active constituents) is very often used. Therefore the present literature search is hoped to pay to intensify research on textile dyes with antibacterial activity. After preparation, number may be assigned as color index for dyed textile.

A diminutive amount of observations are made which may provide inputs for further research.

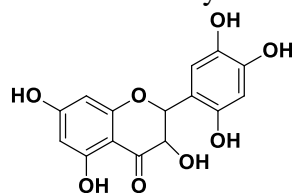
i) Mosquito repellent textiles as recommended by WH.O like DEED, Permethrin (pyrethroid insecticide)...are used in African countries which are used to impregnate the curtains and nets.

ii) At each washing of the textile, use of lemon grass oil or citronella oil may be used while washing. These may repel mosquitoes which may act as antibacterial agent, besides pleasant odor.

iii) These textiles have notorious history as biological weapons. The most widely mentioned red colored heartwood smuggled out of India is red sanders (*Pterocarpus santalinus*). Its water extract provides intense red color (active colored product is called Santaline, Santol may provide red colored textile).

iv) Raw silk is yellow color possibly due to morin which is present in mulberry leaf consumed by silk worm.

vi) R.Rajendran et al., reported the use of natural dye with mordant to color the fabric^{xv}.



Morin

Antibacterial activity of textile fibers with dyes and pigments from marine bacteria is studied by Shivakrishna Pabba et al.,. The results showed that dyed silk fabric material had 50% antibacterial activity. Antibacterial activity of the dyed fiber could be tested by the standard method as given in "Manual Technical manual of the American association of textile chemists and colorists, AAATCC, Research Triangle Park, N.C., 2005, page 80. This method was adopted by Farzanch Alihosseini et al.,^{xvi} to test the antibacterial activity of prodiginine dyed(isolated from marine bacterium) fabrics against *Escherichia cotick-R* or *Staphlococcus aureus*(ATCC 12600). The fabric exhibited antibacterial properties against *E.Coli* and *S.aureus* bacteria after a contacting time of 16 hours. After preparing a dyed fabric number,a test have to be performed which include determination of color strength, better dyeing R/S value

(AATCC manual), wash fastness (AATCC 110106), Permeability of dyed fabric ASTM D737-20, tensile strength -graph test of finished fabrics (ARMD-5034-95-2001), Tear strength (ASTM D-2261-96) and Fabric stiffness.

These antibacterial tests were performed by R. Rajendran^{xvii}. The tests were performed by cotton fabrics dyed with extractive of *Rosaomonafauriae*. This is antimicrobial pink pigment but its active chemical constituent is not known.

Acknowledgement:

The Technical project work comprised in this paper has been carried out under the supervision of A. Radhakrishna, Principal Technical Officer, Department of Computer, Indian Institute of Chemical Technology, CSIR-IICT, Tarnaka, Hyderabad. For suggesting the problem and for his inspiring guidance, constant encouragement by this enlightening discussion throughout the course of this investigation. We express special thanks to Director, IICT, Tarnaka Prof. G. Srimannarayan, (Retd). Professor, Department of chemistry, Osmania University for his constant support, helpful discussion and encouragement. Our sincere thanks to E.V.R. Hanumanth Rao, B.V. Devendra Rao and all senior colleagues in the computer division. I record my sincere thanks to Dr. G.A. Raju, Head, computer division.

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Conclusion:

Paint derived dyes to color cotton fabrics is safe, environmentally, besides they may have antibacterial activity as reported in literature. Therefore promotion of natural dyes given in the chart is strongly suggested.

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Received on July 13, 2023.