

NATURAL DYE: BOON FOR TEXTILE

¹Unnati

ARTICLE DETAILS

Keywords

Natural dyes, synthetic dyes, human being health, textile, environment, mordants

ABSTRACT

Natural dyes have become boon for textile industry after the side effects of synthetic dyes. It was found in different parts of world during 2500-3500 BC. Unlike synthetic dyes it doesn't pollute the environment as well the physical, mental and motor health of human beings. There are different type of natural dyes classified on the basis of origin, application, chemical compounds present in it etc. the dye doesn't fix to the fabric directly, that is why mordants are required to attach the dye to the cloth. Many types of mordants are used, for example- Metallic mordants, Oil mordants, and tannins. This paper reviewed about different sources of natural dyes, their shades, comparison with synthetic dyes and some of the past works of natural dyes on textile.

* Research Scholar, Khwaja Moinuddin Chishti Urdu Arbi- Farsi University, Home Science department, Lucknow, India.

1. Introduction

Natural dyes, dyestuffs and dyeing are as old as textile themselves. Man has always been interested in colours; the art of dyeing has a long past and many of dyes go back into prehistory. It was adept during the Bronze Period in Europe. The primary on paper record of the usage of natural dyes was found in China dated 2600 BC. Dyeing was recognized as early as in the Indus Valley period (2500BC); this knowledge has been substantiated by findings of coloured garments of cloth and traces of madder dye in the ruins of Indus valley civilization at Mohenjodaro and Harappa (3500 BC). Natural material was used to mark hides, adorn shells and feathers, and in cave pictures. Scientists have been able to date the black, white yellow and reddish pigments made from ochre used by primitive men in cave paintings. By the 4th century AD, dyes such as woad, madder, weld, Brazil wood, indigo and a dark reddish purple were known. Brazil was named after the wood found there. Henna was used even before 2500 BC, while saffron is cited in the Bible. The first use of the blue dye, woad by the ancient Briton may have originated in Palestine, where it was found growing wild. The most famed and extremely prized colour through the centuries was Tyrian purple (noted in the Bible) a dye achieved from the spiny dyemurex shellfish. The Phoenicians made it until the seventh century, when Arab victors devastated their dyeing installations in the Levant. In the prehistoric times man used to crush berries to colour mud for his cave paintings. Some of the famous prehistoric dyes include madder, a red dye made from the roots of the *Rubia tinctorum* L., blue indigo from the leaves of *Indigofera tinctoria* L., yellow from the stigmas of the saffron plant (*Crocus sativus* L.) and from turmeric (*Curcuma longa* L.)[1]. Today, dyeing is a complex and specialized science. Almost all dyestuffs are currently made from manmade compounds. This means that prices have been critically reduced and certain use and wear features have been significantly enhanced. However, specialists of the craftsmanship of natural dying (i.e. using naturally occurring sources of dye) uphold that natural dyes have a far higher aesthetic quality, which is much more attractive to the eye. On the other hand, several money-making practitioners feel that natural dyes are non-viable on grounds of both quality and economics. In the West, natural dyeing is now practiced only as a handcraft, while synthetic dyes are being used in all commercial applications. Some craft spinners, weavers and knitters practice natural dyes as a specific feature of their work.

Interestingly, most of the plant materials used for the extraction of dyes form a part of Indian Pharmacopoeia is credited with medicinal properties [2].

There is a huge difference between natural and synthetic dyes. Synthetic dyes have toxicity in nature while natural dyes are nontoxic. With reference to the price synthetic dyes are cheaper than natural dyes because there is less efforts in extraction of dye and more production but natural dyes takes more efforts and less production. Natural dyes degrade easily as compared to synthetic dyes which doesn't degrade and pollute the environment.

2. Types of natural dyes and mordants

Natural dyes can be divided into three categories: natural dyes obtained from plants, animals and minerals. Though some fabrics such as silk and wool can be coloured simply by being dipped in the dye, others such as cotton require a mordant.

2.1 Mordants

Dyes do not interact directly with the materials they are envisioned to colour. Most natural dyes are adjective and need a mordant to fix to the cloth, and stop the dye from either fading with contact to light or washing out. These compounds fix the natural dyes to the fabrics. A mordant is a component which helps the chemical reaction which takes place between the dye and the fibre, so that the dye is engrossed. Vessels used for dyeing must be non-reactive (enamel, stainless steel). Brass, copper or iron pots etc possess their own mordanting qualities. All dyes don't need mordants to help them fix to fabric. If they don't need mordants they are called substantive dyes and if they need a mordant, they are called adjective dyes. Common mordants are alum, iron (or copper), tin and blue vitriol. There are major 3 types of mordants: Metallic mordants: Metal salts of aluminium, chromium, iron, copper and tin; Tannins: Myrobalan and sumach; Oil mordants are mainly used in dyeing Turkey red colour from madder. The main function of the oil mordant is to form a complex with alum used as the main mordant [1].

a) Mettalic mordants: Some important metal mordants are potassium aluminium sulphate, copper sulphate, potassium dichromate, ferrous sulphate, stannous chloride and stannic chloride. Some of the natural dyes can form metal complexes and that is why give different colours. The restrictions to the use of heavy metal salts have been put by the famous 'German Ban'. Accordingly, the suggestive maximum

allowable quantities of different metals in the final product are as follows: As (1.0 ppm), Pb (1.0 ppm), Cd (2.0 ppm), Cr (2.0 ppm), Co (4.0 ppm), Cu (50 ppm), Ni (4.0 ppm) and Zn (20 ppm). The upper limits of the presence of metals differ from product to product and are diverse for different Eco-marks. However, there is no upper limit on iron and tin.

b) Tannins: Tannins are the naturally occurring compounds of high molecular weight (about 500-3000) containing phenolic hydroxyl groups (1-2 per Mw) to enable them to form effective cross-links between protein and other macromolecules. In the dyeing of fabrics, tannins form the basis of so called natural mordant. Tannins are phenolic compounds, and those tannins, which have o-dihydroxy (catechol) groups, can form metal chelates, that give different colour with different metals. An after-treatment with metal salt not only alters the light sorption characteristics of tannins but also makes them insoluble in water. Hence they are fixed on the textile substrate giving good wash fastness.

It is postulated that tannins form the following three types of bond with proteins (e.g. wool and silk) and cellulose (e. g. cotton and viscose rayon).

- Hydrogen bond between the phenolic hydroxyl group of tannins and both the free amino and amido groups of protein or the hydroxyl and carboxyl group of other polymers.
- Ionic bonds between the appropriately charged anionic groups on tannin and cationic groups on protein.
- Covalent bonds formed by the interaction of any quinone or semiquinone group that may be present in the tannins and any other suitable reactive groups in the protein or other polymer.

The vegetable tannins may be distributed structurally into two distinct classes depending on the kind of phenolic nuclei involved and the way they are joined together.

c)

Hydrolysable Tannins: Tannins of this class are characterized by having as a core a polyhydrolic alcohol such as glucose, the hydroxyl groups of which are esterified either partially or wholly by gallic acid or its congeners. Tannins which have this structure can be readily hydrolyzed by acids, bases or enzymes to yield the carbohydrate and a number of isolable crystalline phenolic acids. Thus they are called hydrolysable tannins.

The other acid isolated from hydrolysable tannins is ellagic acid. Therefore, sometimes the hydrolysable tannins of vegetable origin are divided into two groups, namely gallotannins and ellagitannins. Some of the important raw materials for these tannins are: Myrobalan Fruit (*Terminalia chebula*), Oak bark and wood (*Quercus alibi* and other species), Sumac leaves (*Rheas typhoon*), Gallnuts (*Quercus infectoria*) and pomegranate rind (*Punica granatum*).

d) Condensed Tannins: Tannins of this class contain only phenolic nuclei. On treatment with hydrolytic reagents, the tannins of this class tend to polymerise, particularly in acid solutions to yield insoluble often red coloured products known as phlobaphenes. Most tannins of this type are formed by the condensation of two more molecules of flavan-3-ols, such as catechin.

e) Oil mordants: Oil mordants are mainly used in the dyeing of Turkey Red colour from madder. The main function of oil mordant is to form a complex with alum which is used as the main mordant. Since alum is soluble in water and does not have affinity for cotton, it is easily washed out from the treated fabric. The naturally occurring oils contain fatty acids such as palmitic, stearic, oleic and ricinolic etc. and their glycerides. The COOH groups of fatty acids react with metal salts and get converted into COOM, where M denotes the metal.

3. Classification of dyes

Natural dyes can be classified in various ways. Some of the important classifications are:

- i. **Based on origin:** According to origin they are classified into three categories:
 - **Vegetable origin:** Dyes obtained from roots, bark, leaves, trunk, flowers or fruit of the plants for example: turmeric, annatto, henna etc.
 - **Animal origin:** Dyes extracted from insects. Lac, Cochineal and Kermes have been the principal colour yielding insects.
 - **Mineral origin:** Various inorganic metal salts and metal oxides example geru and mineral khakhi. Ocher is a dye obtained from a dirty earthy ore of iron or ferruginous clay, usually red (hematite) or yellow (limonite). In addition to being the principal ore of iron, hematite is an integral part of a number of abrasives and pigments.

ii. **Based on application class:** According to application class, natural dyes are classified as direct dyes, acid dyes, basic dyes, vat dyes and mordant dyes:

- **Direct Dyes:** Turmeric, annatto, harda, pomegranate and safflower are few examples of natural dyes having substantivity for cellulose.
- **Acid Dyes:** These dyes are suitable for dyeing polyamide fibers like wool and silk. Saffron is one of the best known natural acid dye. An after treatment with tannic acid and tartar emetic known as back tanning improves the wash fastness of these dyes.
- **Basic Dyes:** These are cationic dyes used for dyeing polyamide fibres like wool and silk in neutral and mild acidic conditions. These dyes may be applied on cotton mordanted with tannic acid and tartar emetic.
- **Vat Dyes:** The genesis of the name 'Vat Dye' goes to wooden fermentation vessel called vat that was at one time used for reducing the dye in order to convert it into soluble form. Indigo is a well-known natural vat dye for centuries even now used extensively for denim dyeing due to its faded look.
- **Mordant Dyes:** Mordant dyes are defined as those dyes which have affinity for mordanted fibres. All dyes which form complex with mordants, are grouped under this class. Kermes and Cochineal come under this class.

iii. **Based on chemical class:** According to chemical structure they are classified as:

- **Indigoid Dyes:** This is perhaps the most important group of natural dyes. Structure of indigoid dye is shown in Fig. 1. The dyestuff is extracted from *Indigofera tinctoria*, a bush of pea family. The dye is in use since prehistoric times in India. The dyestuff is extracted from leaves of a plant which grows three feet in height and has a maximum dye content of 0.4%. The colouring matter is present in the form of a soluble glucoside known as Indican. Woad is a blue dye similar to indigo, and can be obtained from the fleshy leaves of *Isatis tinctoria*.

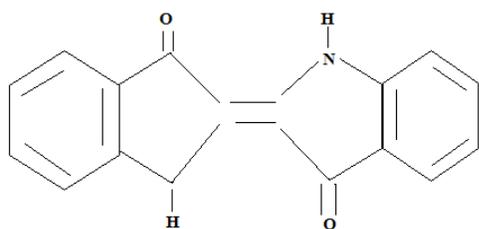
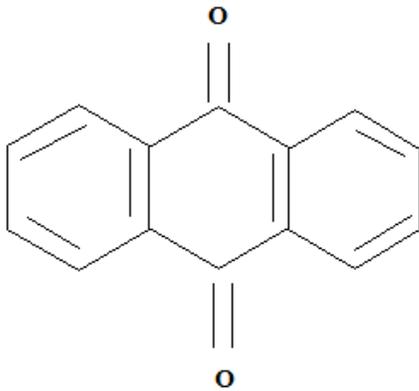
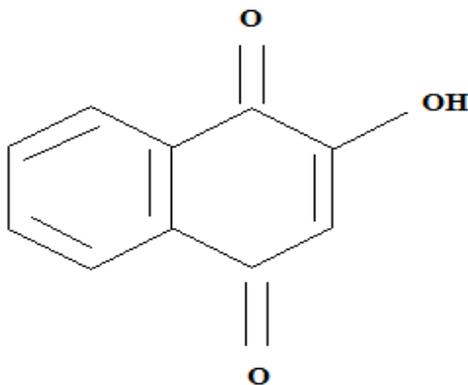


Figure 1: Indigoid structure

- **Anthraquinone dyes:** Some of the most important red dyes are based on the anthraquinone structure (Fig. 2). These are obtained from both plants and insects. These dyes have good fastness to light. They make complexes with metal salts and the result metal–complex dyes have good fastness.

**Figure 2: Anthraquinone structure**

- **Alpha-hydroxy naphthaquinones:** The most prominent member of this class of dye is henna or lawsone or Heena. (*L. inermis* L.) (Fig. 3). Another similar dye is juglone, obtained from the shell of unripe walnuts.

**Figure 3: Alpha-napthaquinones**

- **Flavones:** Most of the natural yellow colours are hydroxy and methoxy derivatives of flavones and isoflavones as shown in Fig. 4.

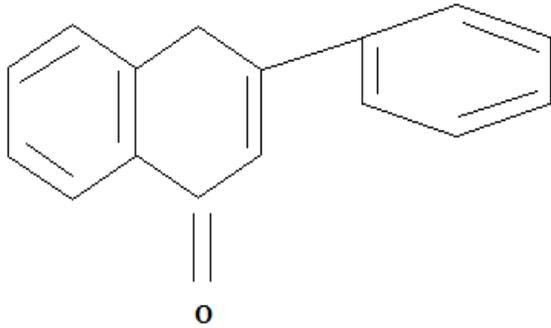


Figure 4: Flavone

- **Dihydropyrans:** Closely related to flavones in chemical structure are substituted dihydropyrans like hematin and its leuco form, hematoxylin. These are the principal colouring bodies of logwood.
 - **Anthocyananidins:** The naturally occurring member of this class includes carajurin obtained from the leaves of *Bignonia chica* Bonpl (cricket wine).
 - **Carotenoids:** The class name is derived from the orange pigment found in carrots. In these the colour is because of the presence of long conjugated double bonds. Typical examples for this group are annatto (*B. orellena*) and saffron.
- iv. **Substantive and adjective dyes.**
- **Substantive dyes:** Dyes which do not require any pre-treatment to the fabric.
 - **Adjective Dyes:** These dyes dye the material only when it is mordant with metallic salts or salt is added to the dyebath (e. g. logwood, madder, cochineal alizarin, etc.).
- v. **Monogenic and Polygenic dyes.**
- **Monogenic Dyes:** These dyes produce only one colour irrespective of the mordant applied along with the dye or present on the fiber.
 - **Polygenic Dyes:** These dyes produce different colours according to the mordant applied e.g. Logwood, alizarin, etc. [3].

Table-1

Sources of different coloured dyes and mordants [1]

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	Solidago grandis DC.	Flower	Alum
Tea	Tectona grandis L.f.	Leaf	Alum
Marigold	Tagetes sp.	Flower	Chrome
Saffron	Crocus sativus L.	Flower	Alum
Tesu	Butea monosperma (Lam)	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

Microorganisms are part of our daily lives. They are small organisms which require a certain medium to grow, such as moisture, temperature, dirt and receptive surfaces. As textile provide the required medium to grow these microbes they can damage the fabrics/garments in several ways, including causing bad odour (on apparel, socks, etc.) leaving stains and reducing the life of product. Apart from these microbes can harm human being by transmitting diseases and infections [4].

Number of Indian herbs do contain antibacterial and antifungal properties. Many common natural dyes reported to have good antimicrobial activity owing to the presence of large amount of tannins. Several other sources of plant which are rich in naphthaquinones, such as henna, walnut and alkanet are reported to exhibit anti-fungal properties [5-7].

4. Past work on natural dyes which can be used to dye textile materials:

4.1. Henna leaves:

Number of Indian herbs do contain antibacterial and antifungal properties. Many common natural dyes reported to have good antimicrobial activity owing to the presence of large amount of tannins. Several other sources of plant which are rich in naphthaquinones, such as henna, walnut and alkanet are reported to exhibit anti-fungal properties [5-7].

As the present trend throughout the world is shifting towards the use of eco-friendly and biodegradable commodities, the demand for natural dyes is increasing day by day [8]. *Lawsonia inermis* L., commonly known as 'Henna' is a shrub or small tree frequently cultivated in India, Pakistan, Egypt, Yemen, Iran and Afghanistan. Powdered leaves of this plant (aqueous paste) are used as a cosmetic for staining hands and hairs [9]. The dyeing property as well as the UV absorption, antibacterial, antispasmodic, corrosion inhibitor were attributed to the presence of lawsone; 2-hydroxy-1,4-naphthaquinone in Henna leaves. This colouring component has following structural formula with Colour Index number 75486 [10, 11].

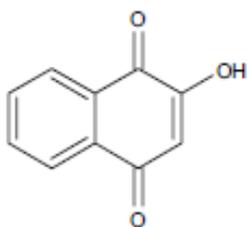


Figure 4: Hydroxy-1,4-naphthaquinone

Plants generally give less amount of colouring component on extraction with water for colouring textiles. Thus yield of natural dyes effects its cost of production that is also one of the factors to restrict the use of natural dyes in comparison with synthetic dyes. According to Ali S et al. his work was undertaken to extract more colouring component with keeping the environment friendly extraction procedure (extraction in aqueous alkaline medium) excluding the extensive application of organic solvents. This medium was chosen to view the acidic chemical nature of lawsone. This will make natural dye as co-partner of synthetic reactive dyes. Furthermore, the dyeing and

mordanting characteristics of colouring matter on cotton have also been studied. Finally, the comparative studies have also been conducted between the dyeings with optimised Henna alkaline extracts and synthetic dyes and evaluated the optimum conditions as well.

Work was on the basic conditions for extraction of natural dye from Henna leaves and the resulting extract was used to further optimize its dyeing conditions on cotton by exhaust method. Dyeings without any mordant were compared with those obtained with premordanting and postmordanting with alum and iron. The alkaline extracts were found to have higher colour than that obtained in distilled water. The washing, rubbing and light fastness properties on cotton, when applied without any mordant, were obtained to be good. In most cases, premordanting and postmordanting with alum and iron did not result in any appreciable increase in fastness properties [12].

4.2. Tea leaves:

Tea plants are classified as *Camellia sinensis* – variety *sinensis* and variety *assamica*. There are six types of teas: green, yellow, dark, white, oolong and black. This classification is based on the processing methods employed, the degree of fermentation and the oxidation of the polyphenols present in tea. The different classes of compounds found in tea include: amino acids, caffeine, carbohydrates, carotenoids, chlorophyll, lipids, minerals, nucleotides, organic acids, polyphenols, saponins, unsaponifiable compounds and volatile compounds [20]. Of the polyphenols, catechins are the principle colorant species. Tsujimura determined the chemical structures of catechins to be (I) epicatechin, (II) epicatechin gallate, (III) epigallocatechin and (IV) epigallocatechin gallate, and named the two gallates as ‘tea tannin I’ and ‘tea tannin II’ respectively [13-15].

Deo H T et al. studied cotton and jute fabrics which were dyed with an aqueous extract of tea, containing tannins as the main colorant species. The dyeing was performed with and without metal salts as mordants, using three different methods named pre-mordanting, meta-mordanting and post-mordanting. The result for wash and light fastnesses of dyed fabrics were good to excellent. The colour of the fabrics was analyzed on computer colour matching system in terms of K/S, and CIELAB colour-difference values. Aqueous extracts of tea yield brown shades with good wash and light fastness on cotton and jute fabrics. Both the light and wash fastness

properties could be further improved by treatment with certain metal salts as mordants. The use of mordants in the dyeing by tea on both the fibers resulted in deeper shades, although with some darkening and dulling. Post mordanting method gave the deepest shades, superior wash and light fastness [16].

4.3. Red calico leaves:

Khan A. A. et al. conducted the study to discover the colouring potential of red calico leaves and to increase colour strength of dye using gamma radiations followed by mordanting process. In this study it is found that the dye can be best extracted from irradiated powder of red calico leaves (RP, 15 kGy) using alkaline medium. Good colour strength is obtained by dyeing irradiated fabric (RC, 15 kGy) with red calico leaf extract at 60 °C for 50 min by maintaining pH of the dye bath at 7.0 using electrolyte concentration of 6 g/L as exhausting agent. It is also found that Copper (1%) and tannic acid (1%) are the best pre and post mordanting agents respectively to improve the colour strength as well as colour fastness properties of irradiated cotton fabric using dye extracted from red calico leaves. Use of gamma radiations particularly, 15 kGy dose can be helpful in improving colourfastness by inducing surface modification of cotton fabric [17].

4.4. Bastard teak:

Carbohydrates, minerals, mucilage, vitamins, and pigments, namely flavonol, crocin, anthrocianin, carotene, lycopene, zigzantin etc. are usually present in petal of flowers. Extensive study is now being done by researchers universally for extraction of colorants from different parts of plant. Most usually accessible raw materials for natural dyes are different parts of the plants which are indispensable for the survival of plants. In India, enormous quantity of herbal materials are unutilized and disposed off daily that can be used for extraction of natural dyes for application in textile industry as a substitute of synthetic dyes. Besides application in textile industry, these dyes can also be used for coloration purpose in food industry, preparation of herbal gulal and manufacture of colorful candles. The primary benefit of natural dyes is that they are cheap, renewable, non-carcinogenic, non-toxic and there is no disposal problems. Some natural sources of dyes can produce truly exquisite shades that are economical to purchase when compared to synthetic dyes. Till date, most of the natural dyed textiles are imported from Third World Countries and India is still a major producer of it.

Sinha K et al. [18] in their research study used natural colorant from the petals of the Flame of forest (*Butea monosperma*) flower was extracted under different operating conditions such as extraction time (45-120 min), temperature (60-90 °C) and mass of the petals (0.5-2 g) by conventional extraction technique. Response surface methodology (RSM) with the help of Design Expert Version 7.1.6 (STAT-EASE Inc., USA) was used for optimisation of the extraction process and evaluation of interaction effects of different operating parameters. The work have shown that natural dye can be successfully extracted from the petals of Flame of forest flower. Maximum dye extraction was observed at 70 °C using 2 g of flower petals. With increasing the extraction time, the dye extraction efficiency increased. Extraction temperature, time and mass of the petal used markedly influenced the dye extraction efficiency from Flame of forest. The optimum conditions were found to be an extraction temperature of 69.72 °C, extraction time of 164.46 min and mass of the petals to be 1.994 g. Under these conditions, the total amount of dye extracted from the Flame of forest petals was estimated to be 9654.5 mg L⁻¹.

4.5. Marigold flower:

An experiment was to study the use of an extract isolated from marigold as a natural dye. The dye potential of the extract was calculated by dyeing, using the flower, in 100 % cotton and silk fabrics under normal dyeing conditions. Studies of the dye ability, wash fastness, light fastness, and colour hue were undertaken. The, L, a and 'b' of materials dyed using the extract were studied with the use of Computer Colour Matching software. The surface colour was not affected by washing, and the quality of the flower was maintained even washing at 60 °C for 30 minutes. Studies have indicated that the change of some of the colors have been noticed after washing with soap. Most of the metal salts exhibited the highest K/S values, due to their ability to form coordination complexes with the dye molecules. These findings reveal that the extract of Marigold flower can be used for coloration of 100 % cotton, silk, and wool fabrics. This article deals with the chemistry, processing, and stability of the pigment and its applications in textile coloration [19].

5. Conclusion

Natural dye is less hazardous than synthetic dyes. It has less or no negative impact on environment unlike man-made dyes. This paper focused on the comparison of synthetic and natural dyes, types of mordants used for fixing of dye, the sources of natural dyes, classification of natural dyes and how some of the dyes were made in the past to dye the textile, the machines used for dyeing, methods used to make those dyes, chemical compound incorporated in the dyes and mordants.

References

1. Gupta D, Jain A and Panwar S, Indian J Fibre Text Res, 30(2005), 190.
2. Dr. Nadiger G S, Sharma K P and Jarag P, Colourage Annual, 2004, 130.
3. Babu K M. Asian Textile Journal, April 2003, 64.
4. Wagner H, Kreher B, Lotter H, Hamburger M O and Cordell, Helv Chim Acta,72(1989), 659.
5. Gerson H, Canadian J Microbiol, 21(1975), 197.
6. Schurech A R and Wehrli W, Eur J Biochem, 84(1978), 197.
7. Bhuyan R, Saikia CN, Das KK. Extraction and identification of colour components from the bark of *Mimusops elengi* and *Terminalia arjuna* and evaluation of their dyeing characteristics on wool. Indian Journal of Fibre & Textile Research December, 2004;29:470–6.
8. Anon. Encyclopedia of chemical technology. 1982;8:315–73.
9. Al-Sehaibani H. Evaluation of extraction of Henna leaves as environmentally friendly corrosion inhibitor for metals. Materialwissenschaft und Werkstofftechnik 2000;31(12):1060–3.
10. Dweck AC. Natural ingredients for colouring and styling. International Journal of Cosmetic Science 2002;24(5):287–302.
11. Ali S, Hussain T, Nawaz R, Optimisation of alkaline extraction of natural dye from Henna leaves and its dyeing on cotton by exhaust method, Journal of Cleaner Production 17 (2009) 61–66.
12. Roy Teranishi and Irwin Horntein, Food Rev. Int., (New York: Marcel Dekker, 1995).
13. M Tsujimura, Nippon Nogeikagaku Kaishi, 10 (1934) 140.
14. M Tsujimura, Sci. Pep. IPCR, 46 (1952) 31.
15. M Tsujimura, Nippon Nogeikagaku Kaishi, 29 (1955) 407.

16. Deo H T, Desai B K, Dyeing of cotton and jute with tea as a natural dye, JSDC 115 (1999) 224-227.
17. Khan A A, Iqbal N, Adeel S, Azeem M, Batool F, Bhatti I A, Extraction of natural dye from red calico leaves: Gamma ray assisted improvements in colour strength and fastness properties, Dyes and Pigments 103 (2014) 50-54.
18. Sinha K, Saha P D, Datta S, Extraction of natural dye from petals of Flame of forest (*Butea monosperma*) flower: Process optimisation using response surface methodology (RSM), Dyes and Pigments, 94 (2012) 212-216.
19. Jothi D, Extraction Of Natural Dyes From African Marigold Flower (*Tagetes Erectal*) For Textile Coloration, AUTEX Research Journal, 8(2) (2008) 49-53.
20. Selvam R. Mari, Singh A.JA. Ranjit, Kalirajan K., " Antifungal activity of different natural dyes against traditional products affected fingal pathogens", vol 18, 1750-1756, 2010.