EXTRACTION OF NATURAL DYES AND MORDANTS FROM SELECTED PLANTS, STUDY ON ITS PIGMENTS AND ITS APPLICATION IN FABRIC DYEING

DISSERTATION SUBMITTED IN PARTIAL FULFILLMENT OF THE

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CERTIFICATE

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This is to certify that this B.Sc. Dissertation entitled 'Extraction of Natural Dyes and Mordants from Selected Plants, Study on its Pigments and its Application in Fabric Dyeing' submitted by Ms. Diya Maria Siril and Ms. Sandra Johnson, Final semester, Bachelor of Science in Botany, Department of Botany and Centre for Research, St. Teresa's College, (Autonomous), Ernakulam is an authentic dissertation work of original investigations carried out by them under my supervision and guidance, and it has not been submitted anywhere else for any other degree, diploma.

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CONTENTS

1. INTRODUCTION

1. INTRODUCTION

Colors are the smiles of nature. Do you think about the earth without colors? We would never want to live in such a world. Colors make the life more aesthetic and fascinating. The colors that directly as well as indirectly associated with the maintenance of life on earth. The colors are associated with emotions, human qualities, seasons, festivals and passion in our life. Two important phenomena where the involvement of color is indispensible are photosynthesis and pollination. In photosynthesis green plants capture light energy and convert it into chemical energy. This is the precursor step in any process for obtaining food for any organism and capability of green plant to capture light energy is due to the presence of pigment molecules called, chlorophyll and another important process is the pollination, it is one of the fundamental process not only for the pollen dispersal and the creation of new individuals of plants but also for the survival of pollinating agents. Most of the angiosperm plants are animal pollinated and the pollinators are attracted towards the flowers due to the presence of various colors in their petals. This is mainly due to the presence of numerous pigment molecules. The various types of pigment molecules are one of the products gifted by nature for various purposes. In the human civilization, the plants not only used for basic needs of life such as food, fuel, fiber and cloth but also they are the rich source of natural dyes for various purposes. They are rich source of vivid colors ranging from yellow to black.

India is one of the 17^{th} mega diverse countries in the world, houses wide and diverse classes of natural products including various sources of dyes or pigments. The dyes have the ability to bind to a substrate but the pigments, no such ability. Natural dyes are the dyes or colorants that are derived from plant, animal or mineral sources. Majority of the natural dyes are obtained from plant sources such as: - roots, berries, leaves bark and wood. Natural dyes are one of the renewable and sustainable bio resources and it is used in textile as well as food industry with minimum environmental and health impacts. Archaeological evidences suggest that the art of dyeing was old as human civilization and it is dating back to the Neolithic period. In Ayurveda, there are some descriptions about the natural dyes and the historical evidence suggest that the use of natural dyes in wall paintings of Agenda, Ellora and Sithanvasal and they still prove their efficacy of dying craft and that had been inherited from ancient times in India. The other traces of the using natural dyes were also found in China in the year 2600 BC. Later, colored pigments

were found in Egypt in the tomb of King Tutankhamun. Alexander the Great mentioned purple clothing in 541 BC.

In the textile industry, some of the dyes are used alone because they will give good color when fixed in the textile fiber they are called direct dyes. But some of the dyes require a mordant to fix the color in the textile fiber such dyes are called adjective dyes. A mordant is an element that can enhance the chemical reaction between textile fiber and the dye as a result the dye can easily fixed into the textile material and it can also use to increase the color fastness. One of the advantages of natural dye is that it is eco-friendly and biodegradable, so they do not pollute the environment by the accumulation of unwanted waste chemicals. The increased development of Science and Technologies and decreased awareness of environmental protection leads to the discovery and popularization of synthetic dyes. But with the advent of synthetic dyes environmental pollution increased day by day. The man made dyes are badly affecting the aquatic organisms by destructing its photosynthetic activity. Some of the dyes are allergic, carcinogenic and mutagenic too. 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 its dreadful and silent killing effects. In India, Thirupur (Tamil Nadu) known as the "Banyan city of India" acute soil and water pollution occurring due to effluents from dying units. Effluents from dyeing units in the area heavily polluted river and land in the area thus State Government has imposed stringent restriction to function such dying units and also ordered to shut down many of dyeing units those who violate the norms of pollution control board. This situation occurring in most of the textile hubs all over the World lead to recovery and rediscovery of traditional natural dyes. Recently, interest in the use of natural dyes has been heightened due to the result of stringent environmental standards imposed by many countries in response to toxic and allergic reactions associated with synthetic dyes. The World Health Organization and USFDA specified and set definite control limits for the use of synthetic colors, because of their serious toxic reactions towards eyes and skin. Possibly this may be the fact of recent awareness and resurgence of interest all over the world towards natural resources for getting biodegradable and environmental friendly dyes.

Now it is the era of Green Minded People, because most of the natural dyes possess health promoting and Eco-friendly properties. Now the increased awareness of environmental

protection and hazardous effect of synthetic dyes can again popularize the use of non-toxic, non-allergic and eco-friendly natural dyes.

Types of Natural Dyes

Based on the source of natural dyes are obtained, it can be classified into three types. Natural dyes obtained from – 1) Plants 2) animals and 3) minerals

1) **Natural dyes from plants:**

Many natural dyestuff and stains were obtained mainly from plants dominated as a source of a natural dyes, producing different colors like red, yellow, blue, black, brown, and combination of these. Almost all parts of the plant like- roots, berries, bark, leaves and wood, seed, flower etc. produces dyes. About 2000 plants produce dyes and out of which 150 have been exploited for various dyeing purposes, In India more than 450 taxa are known to produce dyes.

2) **Natural dyes from animals**

Mollusc, murex snail, cuttlefish, shell fish, insects etc. are used as an important source of natural dye. Cochineal and lac are the dyes obtained from insects. Cochineal dyes are red dyes which are obtained from the insects living on cactus plants. The properties of cochineal bugs are first discovered by pre Columbian Indian and this dye is still used in some places.

3) **Natural dyes from minerals**

Mineral origin dyes are derived from specific mineral natural resources or they are derived from purified inorganic compounds. Clay, ochre, malachite, chrome –yellow, iron buff, narkin yellow etc. are some of the mineral dyes. These dyes are widely used in several ancient paintings and buildings. Ochre is an important one, which are obtained from an impure earthy ore of iron or ferruginous clay, usually red (hematite) or yellow (limonite).

Natural dye resources used in present study

1. ***Beta vulgaris*** **(Family: Chenopodiaceae; Common name: Beetroot, sugar beet)**

   Beetroot is a biennial herb with stems up to 1.25 m height. Leaves simple, petiolate, ovate or oblong become linear bracts in the inflorescence. Flowers are small, numerous, greenish, clustered in a large and diffuse terminal panicle. The parts used are roots. It is

native of coastal areas of Europe, North Africa, some parts of Asia including Indian subcontinent India. This root tuber cultivated in subtropical climates all over the world. The beetroot is the tap root portion of a beet plant. The beet produces large, succulent, fleshy sugary roots, a short crown stem, and a rosette or many long-stalked leaves during the first year. During the second year, it produces flowers and seeds. After the seed crop is produced, the entire plant dies. The leaves are glabrous, ovate, dark green, or red, frequently forming a rosette from the underground stem; and the roots are conspicuously swollen at the junction with stem. The taproots are white, fleshy; deep penetrating. Usually, the deep purple roots of beetroot are eaten boiled, roasted, or raw, and either alone or combined with any salad vegetable. The green, leafy portion of the beetroot is also edible.

2. ***Caesalpinia sappan*L.(Family: Fabaceae; Common name: Sappan wood, Brazilian wood tree)**

   Sappan wood is a small thorny tree, 6-9 m in height and 15-25 cm in trunk diameter with a few prickly branches. Leaves are double-compound alternately arranged, with 8-16 pairs of side-stalks. Side-stalks are prickles at the base and with 10-20 pairs of oblong, leaflets, very oblique at base, rounded to notched at the tip. Yellow flowers are borne as panicles in leaf axils and at the end of branches. Flowers are fragrant, pentamerous. Stamens are waxy-white, filaments densely woolly at the base. Fruits are woody pods, compressed with a hard recurved short beak, with 3-4 seeds. The heartwood which is used in medicine is light yellow when freshly cut, but it quickly changes to red. The color diffuses out easily in hot water.

3. ***Rubia cordifolia* L. (Family: Rubiaceae; Common name: Manjistha, Indian madder)**

   Manjishtha is a perennial, herbaceous climber with very long, cylindrical roots having a thin red bark. They can grow up to 1.5m in height. Stem is long, rough, slightly woody at the base, quadrangular, and glabrous. Branches are climbing means of numerous prickles. Leaves are heart shaped, five-nerved from the leaf base, and occur in whorls of four. Petiole is roughly triangular with many sharp recurved prickles on the edges. Flowers are small, yellow, and scaly, and occur in terminal cymes. Fruits are globose, smooth, shining, violet or purple black in color with grey black seeds. The species is found

throughout the hilly subtropical to sub-temperate regions of India, between 300 m and 2000 m altitudes.

Mordants

Mordants are elements which create a bonding between dye and fabric. They can facilitate a chemical reaction between textile fibre and dye particles or pigment mordants. Mordants not only give the dye an affinity, but in many cases, they produce different colors and improve the fastness of a dye. There are many plants which may yield a color that is brilliant and pleasing but fades easily, unless fixed by using a mordant .Several different methods of applying a mordant are in use, the principal methods are as follows;

1. Pre-mordanting - Applying the mordant before the dye
2. Simultaneous mordanting - Applying both together
3. Post mordanting - Applying the mordant after the dye

   In the present project work pre-mordanting method was used.

Types of mordants:

Mordants can be classified into three types (1) Metallic mordants (2) Oil mordants (3) Natural mordants. Metallic mordants are metal salts of chromium, ion, and aluminum, copper and tin. Oil mordants are Turkey red oil and natural mordants are tea leaves, myrobalan and vinegar.

1. ***Phyllanthu semblica*** **(Family: Phyllanthaceae; Common name: Indian Gooseberry, Amla)**

The tree is small to medium in size, reaching 1-8 m in height. The branchlets are not glabrous or finely pubescent, 10–20 cm long, usually deciduous; the leaves are simple, sub-sessile and closely set along branchlets, light green, resembling pinnate leaves. The flowers are greenish-yellow. The fruit is nearly spherical, light greenish-yellow, quite smooth and hard on appearance, with six vertical stripes or furrows. The fruit is up to 26 mm in diameter, and, while the fruit of wild plants weigh approximately 5.5g. The fruit of Amla is rich in vitamin C (ascorbic acid) and contains several bioactive

phytochemicals, of which majority are of polyphenols such as ellagic acid, chebulinic acid, gallic acid, chebulagic acid, tannins, quercetin, corilagin, leutolin, etc.Sugar-substituted phenolics as well as tannins are reported in fruit's pulp. Economic importance includes; fruit sour and astringent, cooling, diuretic, laxative, eaten raw or cooked, also pickled, a rich source of vitamin C.

2. ***Terminalia chebula* Retz. (Family: Combretaceae; Common name: Myrobalan)**

It is a deciduous tree, to 25 m high, bark 5-6 mm thick, surface dark brown to black, fissure shallow, vertical, exfoliating in thick scales; blaze yellowish –brown; young shoots densely pubescent; branch lets brownish or greyish, glabrous.Leaves are simple, opposite to alternate, ex- stipulate; petiole 12-25 mm long. Flowers are bisexual, greenish-white, 5-6 mm across, in terminal and axillary spikes with offensive smell. Fruit is a drupe, obovoid, woody, 5 angled, glabrous, greenish–yellow and one seed is present. The main components present in the fruit pulp are; hydrolysable tannins, phenolic carboxylic acid like gallic acid, ellagic acid, chebulic acid and gallo tannins such as 3,4,6 tri-o-galloyl-β-D-glucose,1,6 di-o-galloyl-β-D-glucose etc. Ellagitannnin, chebulin and phenolic compounds (Rathinamoorthi et al., 2014) are present in *T. chebula* fruits.

3. ***Averrhoa carambola* (Family: Oxalidaceae; Common name: Star fruit, carambola)**

Carambola, also known as star fruit, is the fruit of *Averrhoa carambola*, a species of tree native to tropical Southeast Asia. The mildly poisonous fruit is commonly consumed in parts of Brazil, Southeast Asia, South Asia, South Pacific, parts of East Asia, the United States, and the Caribbean and contains the neurotoxin. The tree is cultivated throughout tropical areas of the world. The fruit has distinctive ridges running down its sides (usually 5–6).When cut in cross-section; it resembles a star, giving it the name of starfruit. The entire fruit is edible, usually raw, and may be cooked or used as preservative, as garnish, and juices. Generally, star fruits contains following components, natural phytochemicals such as flavonoids, terpenes, saponins, alkaloids, proanthocyanins, vitamin C, tartaric acid, oxalic acid, α-ketoglutaric acid, citric acid, vitamin B1 and B2, carotene, cellulose,

pectin, gallic acid, fatty acids, fibers, volatile flavors, hemicelluloses, polysaccharides, and sterols.

Objectives of the study

- ❖ To evaluate locally available natural dye resources for textile dyeing.
- ❖ To examine use of natural mordants for textile dyeing using various natural dyes.
- ❖ To identify most suitable dye-mordant combinations.
- ❖ To explore possibility to produce various dye shades using combinations.

2. REVIEW OF LITERATURE

2. REVIEW OF LITERATURE

In the modern era, widespread emphasis is given to the utilization of natural dyes as it is a safe and eco-friendly resource for the sustenance of humans. Though there are some problems associated with the use of natural dyes, natural dyes have regained the attention of the world. There are reports that certain natural dyes can impart antimicrobial properties to the dyed fabric (Thilagavathi et al., 2005). Switching over to natural dye resources like plants can lead to a healthy way of living with sustenance (Kadolph, 2008).

The use of natural dyes dates back to ancient periods and continued as coloring source until the turn of the twentieth century (Kharbade and Agrawal, 1988; Gilbert and Cooke, 2001). Even though the earliest dyes were discovered accidentally using berries and fruits, with experimentation and gradual development, the art of dyeing using natural dyes was perfected in ancient civilizations (Kharbade and Agrawal, 1988; Mishra and Patni, 2001). However, with industrialization at the turn of the 20^{th} century, natural dyes were mostly replaced with its synthetic alternatives. Madder (*Rubia cordifolia*) is such a neglected red dye once largely been used as a textile dye that gives wide and diverse shades of red, indigo, or even black (Devipriya, 2018). The source of Indian madder is the mature roots of *R. cordifolia*. Root-derived powder of madder was an important dye source for the Asian cotton industry and is still used by craft dyers in Nepal (Gulrajani, 2001). The madder dye imparts brilliant colors of red depending on the mordants and dye concentrations. Depending on the mordant used, the color shade of madder dye can be modified through red, pink, orange, lilac, and brown (Green, 1995).

In India, there are more than 450 plants that can yield dyes. In addition to their dye-yielding characteristics, some of these plants also possess medicinal value (Siva, 2007). Natural dyes are environment friendly for example, turmeric, the brightest of naturally occurring yellow dyes, is a powerful antiseptic that revitalizes the skin, while indigo gives a cooling sensation. (Chengaiah et al., 2010). *Punica granatum* L. and many other common natural dyes are reported as potent antimicrobial agents owing to the presence of a large amount of tannins (Hussein, 1997). Several other sources of plant dyes rich in naphthoquinones, such as lawsone from

Lawsonia inermis L.(henna), juglone from walnut, and lapachol from alkanet, are reported to exhibit antibacterial and antifungal activity (Singh et al., 2005; Chengaiah et al., 2010).

Natural dyes: past, present and future

Indians are forerunners in the art of natural dyeing (Vankar, 2000). An early understanding of dyeing techniques and their applications was empirical and was not backed by scientific reasoning (Cardon, 2009). Age-old documents on the use of curcumin and madder as yellow and red natural dyes are available (El-Shishtawy et al., 2009). Primitive dyeing techniques included sticking plants to fabric or rubbing crushed pigments into cloth (Quye et al., 2004). The methods became more sophisticated with time and techniques using natural dyes from crushed fruits, berries, and other plants parts, which were boiled into the fabrics and water fastness, were developed (Vanker, 2000). It is always useful and interesting to test the dye or fabric to be dyed. The outcome will depend on the fabric, the mordant that has been used, and the dye that has been chosen (Kapoor, 2006). The physical properties of natural dyes were investigated in the past (Acquaviva et al., 2009). The dying methods by applying modern techniques including nano techniques are reported (Acquaviva et al., 2009).

In India, with its diversity of climatic zones and altitudes, geographic variety has resulted in a rich biodiversity that has gifted flora and fauna, which yields an array of dye-producing shrubs and perennials. The ancient people exclusively used dyestuffs of vegetables, minerals, and animal origin, all easily obtained in their own vicinity. In India, it was widely used for coloring of fabrics and other materials (Kapoor 2006). In order to meet the growing demand for natural colorants, new pigment crops are being sought. The newer sources need to be tapped for the purpose. However, for some sources, such as roots of manjistha (Rubia *tinctoria*), safflower (*Carthamus tinctorius*), and indigo (*Indigofera tinctoria*), there is organized cultivation because of their exceptional and established versatility. Increased acreage is devoted to root and safflower cultivation purely because of their dye qualities. More plants need to be considered as agronomically viable plantations. Natural dyes per se are sustainable as they are renewable and biodegradable. The experience with natural dyeing has given an insight into exploring plants in the neighborhood. Finding fiber colors in plants that grow easily and fast invites the attention of the new world of fiber colors that give exotic shades. These natural colors have richness and

luster that synthetics can never attain. It has become a common misconception that natural dyes only produce beiges and browns, and other washed-out shades. In reality, vibrant, fast, natural colors can be produced, which are comparable with and often surpass the colors of synthetics. Apart from the sources of these dyes, it is perhaps the commitment of those propagating them and the near clinical efficiency with which dye is extracted, produced, and used which is responsible for the unique nature of natural dyeing and producing stable coloration (Siva 2007). Modern cultivation system for getting maximal dye yields, including optimal seeding and harvesting time and optimal fertilization procedures, is to be adopted to maximize the dye and biomass yield (Kapoor 2006). The utilizable plant parts were collected and subjected to specific dehydration processes, or the dyestuff was extracted as per the standardized strategy. It will help in giving an idea about the feasibility of providing high-quality natural dyes from plants, creating new opportunities for both farmers and the fabric industry in line with the current consumer trends toward ethnic fabric and natural products. Several newer sources of natural dyes, particularly for dyeing cotton and silk fabrics to get a gamut of colors have been explored. This will increase the availability of newer shades and new choices of fabric dyed with natural dyes (Vankar, 2017).

In the past, extensive surveys and documentation of various natural dye resources for fabric dying were carried out in different parts of the world; India (Mohanty et al., 1987; Chandramouli, 1995; Siva, 2007), North East India (Bhuyan and Saikia, 2005; Mahanta and Tiwari, 2005), Assam, India (Kar and Borthakaur, 2008), Uttrakhand, India (Gaur, 2008), Turkey (Ozgokce et al., 2003; Dogan et al., 2004), and Thailand (Moeyes, 1993).

The use of several plants species was explored and their potential as a source of fabric dye was extensively studied, e.g., *Rubia cordifolia* (Singh et al., 1993; Vankar et al., 2008; El-Shishtawy, 2009; Devipriya and Siril, 2022), *R. tinctorum* (Angelini et al., 1997), *Prunus persica* (Sandeep et al., 2005), Gulmohar- *Delonix regia* (Anitha and Prasad, 2007; Purohit et al., 2007), eriophyid leaf galls of *Quercus leucotrichophora* (Mishra and Patni, 2001), *Eleutherine bulbosa* (Manjula and Bindu, 2022), *Acacia* sp. (Ratnapandian et al., 2012). *Garcinia mangostana* (Chairat et al., 2007; Chantrapromma et al., 2007), Sappan wood- *Caesalpinia sappan* (Ghorpade et al., 2000; Lee et al., 2008; Mitsunaga et al., 2009), *Eucalyptus* leaves (Weiner et

al., 2010). Flowers of *Canna indica* (Srivastava et al., 2008), *Salvia splendens* (Vankar and Kushwaha, 2011), *Rhododendron* sp. (Vankar, 2010) were used for the dyeing of cotton and silk fabrics. *Acacia arabica* bark-based natural dye was used to dye cotton fabrics (Tiwari et al., 2001). The use of henna (*Lawsonia inermis*) leaves, Heartwood of Catechu (*Acacia catechu*), and bark of Babool (*Vachellia nilotica*) for the textile dyeing was reported elsewhere (Vankar, 2002). Tea (*Camellia sinensis*) leaves were reported as a natural dye source (Deo and Desai, 1999).

Apart from studying the use of plant-based extract for fabric dyeing, detailed chemistry of natural dyes was worked out in the past (Vanker 2000, 2017). For fabric dying, wide categories of phytoconstituents were identified in the past (Bhat et al., 2005). Physical and chemical investigations on natural dyes were accomplished (Acquaviva et al., 2009). Vankar et al. (2008) developed sonicator-based dyeing of cotton, wool, and silk using *Mahonia napaulensis* fruit-based extract.

Reports on the use of natural dyes to dye various types of fabrics are available. For instance, methods for cotton fabrics dying using different plant-derived dyes are available (Devipriya, 2018). Eom et al. (2001) used cotton dyeing using the cationization method. Cotton fabrics were dyed using the golden drop dye method (Sandeep et al., 2003). Methods for the dyeing of modified acrylic fibers were also reported (El-Shishtawy, 2009). Singh et al. (1993) optimized the procedure for dyeing silk with natural dye extracted from the roots of *Rubia cordifolia*. Ultrasound-energized dyeing of cotton fabric was tested using canna (*Canna indica*) flower extracts (Ghorpade et al., 2000). Kale et al. (2006) have described the utilization of *Cosmos sulphureus* Cav. flower dye on wool fabric. Tiwari et al. (2000) forwarded ultrasound dyeing with *Ocimum sanctum* using eco-friendly mordants.

The effective binding of dyestuff to the fabric requires suitable mordants. A wide variety of synthetic or natural mordants are applied in conjunction with natural dyes. The use of such a combination often provides stable dyeing and essentially provides varying shades of the original dyestuff or vibrant color shade different from the basic color shade of the dye (Garg et al., 1991). Use of metal ions as mordant was reported earlier (Zhu and Sibiao, 1991).

Study on pigments

Beta vulgaris

Betanin is the water soluble chief red glycosidic pigment of beetroot, and it is the only betalain approved for use in food, textile and pharmaceutical products as a natural red colorant. In fabric, beet dye is permanent, but it may fade a bit with a few washes. Beets will barely dye cottons but will yield a lovely, subtle, peachy-brown tone on wools. Using a fixative, like mordant, can help it take better. Betanin degrades when subjected to light, heat, and oxygen. The stability of betanin color is pH sensitive and generally is less than that of artificial dyes. The color of betanin is most stable between pH 4.0 and 6.0. Thermostability of betanin was pH dependent and was greatest between 4.0 and 5.0. Betanin also has anti-inflammatory effects and protects hepatic functions in humans.

Caesalpinia sappan

Brazilin is the major component isolated from the heartwood of *C. sappan.* Brazilin is an organic compound that is a red pigment obtained from the wood of sappan. It has a role as a plant metabolite, a histological dye, an antineoplastic agent, a biological pigment, an anti-inflammatory agent, an apoptosis inducer, an antioxidant, and an antibacterial agent. Brazilin is also used as a natural red pigment for histological staining.

Rubia cordifolia

Major coloring components of madder (Rubia cordifolia) root: (a) purpurin (b) munjistin (c) alizarin. Alizarin and purpurin are two main anthraquinone-type colorants found in the root and tubers of *R .cordifolia* (Indian Madder) alizarin and purpurin have antimicrobial and antifungal activity against different pathogenic bacteria (Bechtold andMussak, 2009; Chenciner, 2000). Purpurin even reveals an antigenotoxic effect against a range of different environmental carcinogens. In medicinal application, madder has been used in skin care products with astringent, tonic, and antiseptic functions. It can be used to clean open wounds and treat skin diseases.

The brief survey and review of the literature point out that the utilization of natural dye resources in fabric dyeing has got wide attention in the past. Several new plant-based dyes, new mordant dye combinations, novel dyeing methods, and the identification of pigment chemical strictures are some of the prominent areas of research in this field.

3. MATERIALS AND METHODS

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3.1. Fabric dyeing

3.1.1. Plant material used for dye preparation

- The tuber of *Beta vulgaris*,
- The wood shavings of *Caesalpinia sappan*,
- The dried roots of *Rubia cordifolia*

3.1.2. Extraction of crude dye

Based on nature of plant material the procedure for the extraction of dye from plant part slightly varied from the standard protocol. The plant materials were dried, powdered and weighed out (100 gm) and the dye was eluted by using water bath. The powdered plant material was taken in a beaker (2 litre) and it was then dissolved in 800 ml of distilled water and heated in a water bath for 1 hour at 90°C.The resulting solution was left overnight for sedimentation. The clear dye solution obtained was carefully poured into a conical flask (1 litre) and then filtered through a fine sieve to remove fine debris. Using a rotary evaporator (60°C) water content was evaporated from the exudates.

3.1.2.1. *Beta vulgaris*

For the preparation of the dye, the root tuber of *Beta vulgaris* was collected from a vegetable vendor in Varappuzha. The tuber was cut into small pieces (approx.2x2 cm size) and then grind in a mixer grinder. Then 100g of the paste was weighed out into a beaker and 50ml of distilled water was added, and the paste was diluted to 500ml by using water. After that, the solution was kept for 2 hours and then it was filtered by using double-layered cloth and again filtered by using a sieve for the complete removal of tuber debris from the solution.

3.1.2.2.*Caesalpinia sappan*

The dried wood shavings of sappan wood were bought from a grocery shop in Aroor. It was then powdered using a mixer grinder. Then 100 g of powder was mixed in 600ml of distilled water and it was then heated in a water bath for 2 hours at 90°C. After 2hr, the dye solution was filtered through double layered cloth and repeated by using a fine sieve to remove debris from it.

3.1.2.3. *Rubia cordifolia*

The dried roots of *Rubia cordifolia* was bought from an Ayurverdic drug shop, Varapuzha. It was then powdered using a mixer grinder. Then 100 g of the powder was taken, to which 600ml of distilled water was added. It was then kept in water bath for 2hr at 90^{o}C. The dye solution was then kept undisturbed overnight. It was then filtered using a cheese cloth. It was then again kept undisturbed, allowing it to sediment and then carefully decanted.

3.2. Plant material used for mordant preparation

- The fruits of *Phyllanthus emblica*
- The seeds of *Terminalia chebula*
- The fruits of *Averrhoa carambola*

3.2.1. Preparation of mordant solutions

Mordanting is an important process in textile dyeing. A mordant is not only used to fix the dye to the fabric but also to increase the color fastness. Based on different mordant the same dye may darken, brighten or drastically alter the final color of the dyed fiber (Cho et al., 2008; Manhita et al., 2011). For the preparation of mordant solution, the plant material was dried and powdered by using mixture grinder. The powder (1:100) was soaked in tap water for 12 h at room temperature and after that the solution was mixed with 100ml of water and allowed to heat in a water bath for 30 minutes at 80°C. Then the mordant solution was filtered by using single layered muslin cloth. After the recovery of mordant it was used for dyeing experiments.

3.3. Processing of the fabrics

Pure, white, 100% cotton fabric purchased from the local market (Varapuzha, ErnakulamDistt.) was soaked in water for 24 hours and it was rinsed several times with water to facilitate destarching. It helps the adsorption and penetration of applied dye and also helps in the even spread of the dye. The shade dried cloth was then used for the dyeing experiment.

3.4. Dyeing of cotton clothes

The cotton clothes of 20cm×20cm size were taken. The scoured cotton fabric was treated with different mordanting solution using pre mordanting method, the mordant was applied first and it is followed by dyeing the sample. The dye bath was taken in a beaker and heated in water bath. Temperature was maintained at 95°c for 45 minutes. The material to dye liquor ratio was fixed as 1:20. After the mordanting and dyeing, the dyed material was repeatedly rinsed with water for removing excessive dye and mordant. The OD and reflectance were measured by using UV- Vis spectrophotometer.

3.4.1. Determination of absorption wavelength and absorbance by the addition of different mordants

By using UV-Vis spectrophotometer, the absorption maxima of each dye extract was identified. On the basis of spectral record, wavelength was fixed for recording effluent OD. Most suitable dye - mordant combination was determined by least OD value as the minimal release of dye stuff to effluent.

3.4.2. Analysis of color fastness

Color fastness was calculated by washing the cloths with water after dyeing in two or three times. The OD value was determined after each washing with the help of UV-Vis spectrophotometer. Based on OD color fastness was determined.

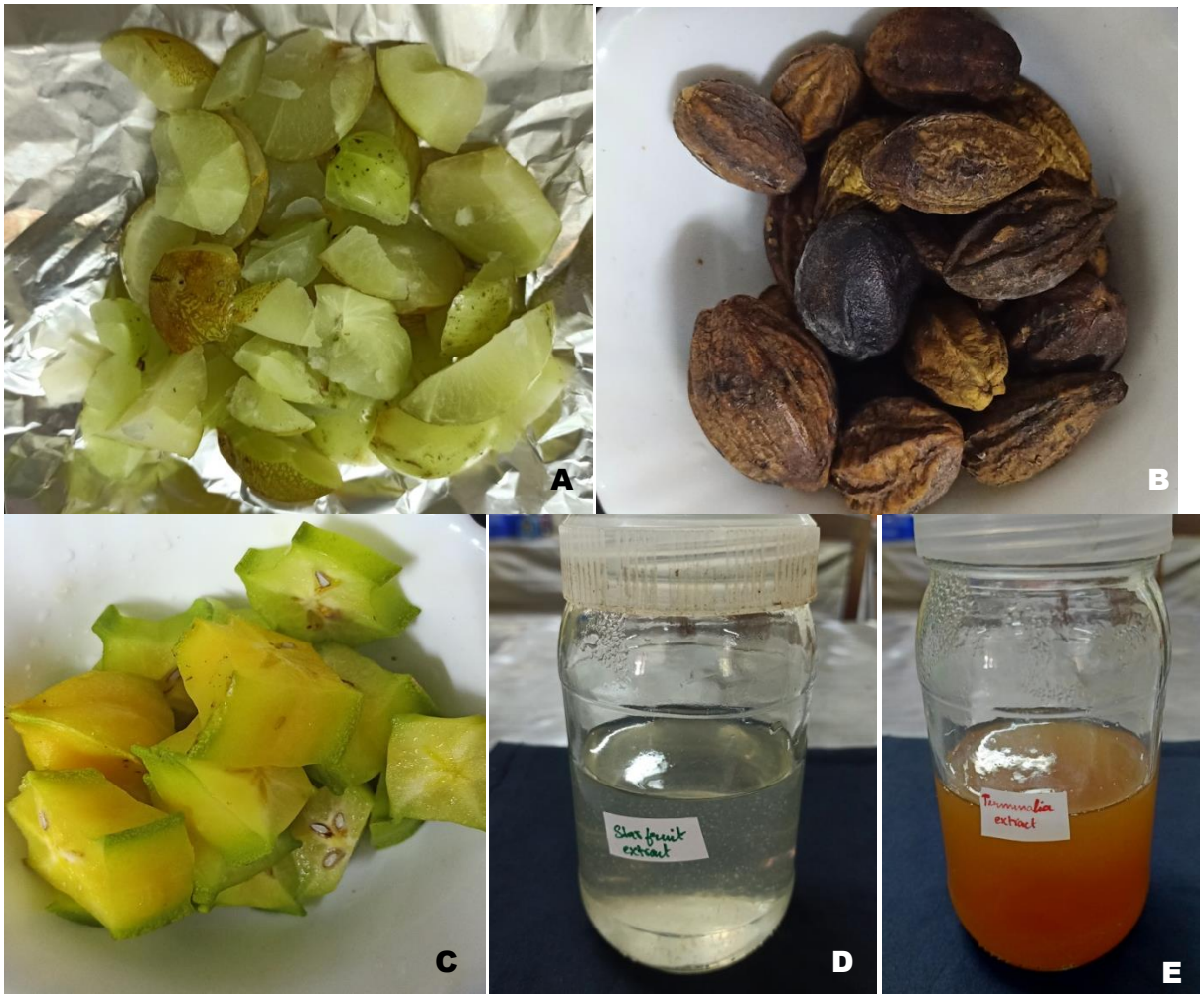

Fig 1: Mordants used in the fabric dyeing;
A –*Phyllanthus embelica* (Goose berry),, B – *Terminalia chebula*(Myrobalan), C –*Averrhoa carambola* (Star fruit), D – *Averrhoa carambola* extract, E – *Terminalia chebula* extract, F – *Phyllanthus embelica* extract.

Fig 2: Plant materials used to prepare dye extract solution; A – *Beta vulgaris* (Beetroot), B – *Caesalpinia sappan* (Sappan wood), C – *Rubia cordifolia* (Indian madder).

4. OBSERVATIONS& RESULTS

4. OBSERVATIONS AND RESULTS

4.1 Fabric dyeing using *Beta vulgaris* tuber extracts

Fabrics dyed with beetroot dye using various mordanting method showed varying shades red pink color (Figure 4.1). Least effluent OD (1.52) value was recorded in *Terminalia chebula* based mordanting. Highest OD value (Table 4.1) was recorded when *Averrhoa carambola* used as mordant. For determining color fastness, OD value was recorded in each washing. After the third washing the least OD (0.100) value was obtained when *Terminalia chebula* based mordanting was done.

Table 4.1. Dyeing property of *B. vulgaris* dye extract on cotton cloths in presence of different types of mordants.

Mordant used	OD of the beetroot dye before dyeing	After dyeing
Phyllanthus emblica	2.18	1.67
Terminalia chebula	2.18	1.52
Averrhoa carambola	2.18	1.93

Table 4.2. Color fastness of *B. vulgaris* dyed cotton fabric in consecutive washing recorded by analysing OD of effluent from first three washing.

Mordant used	After 1st washing	After 2nd washing	After 3rd washing
Phyllanthus emblica	0.116	0.102	0.103
Terminalia chebula	0.115	0.104	0.100
Averrhoa carambola	0.132	0.126	0.113

4.2 Fabric dyeing using the dried bark of *Caesalpinia sappan*

In the presence of different mordant solution the bark extract showed varying shades of red orange color (Figure 4.2). Cotton fabrics dyed using *C. sappan* showed orange shades with varying intensities. Spectrophotometric analysis of dye effluent after first washing showed highest OD value in the case of *A. carambola* based dyeing and least OD value was noted in the case of *T. chebula* (Table 4.3).

Table 4.3 Dyeing property of *C. sappan* dye extract on cotton cloths in presence of different types of mordants.

Mordant used	OD of the sappan dye before dyeing	After dyeing
Phyllanthusembelica	2.26	1.12
Terminaliachebula	2.26	1.63
Averrhoacarambola	2.26	2.10

Table 4.4. Color fastness of C.*sappan* dyed cotton fabric in consecutive washing recorded by analyzing OD of effluent from first three washing

Mordant used	After 1st washing	After 2nd washing	After 3rd washing
Phyllanthusembelica	0.118	0.92	0.014
Terminaliachebula	0.106	0.056	0.023
Averrhoacarambola	0.127	0.117	0.075

Subsequent washing showed a reduction in the OD value (Table 4.4). When *P.emblica* was used as mordant in the method of dyeing, OD declined to 0.014 levels.

4.3 Fabric dyeing using the dried roots of *Rubia cordifolia*

The cloth dyed with the *R. cordifolia* dye in presence of different mordants showed varying shades of brown color (Figure 4.3). Highest OD (1.93) was recorded in the case of *A. Carambola* mordanting. In presence of *T. chebula* mordanting, least OD (1.21) value was achieved. The result indicates the good binding property of the dye due to the presence of *T .chebula* extract on the fabric.

Table 4.5. Dyeing property of *R. cordifolia* dye extract on cotton cloths in presence of different types of mordants.

Mordant used	OD of the *R. cordifolia*dye before dyeing	After dyeing
Phyllanthus emblica	2.83	1.44
Terminalia chebula	2.83	1.21
Averrhoa carambola	2.83	1.93

Table 4.6. Color fastness of *R. cordifolia* dyed cotton fabric in consecutive washing recorded by analyzing OD of effluent from first three washing

Mordant used	After 1st washing	After 2nd washing	After 3rd washing
Phyllanthus emblica	0.163	0.100	0.054
Terminalia chebula	0.102	0.088	0.032
Averrhoa carambola	0.123	0.093	0.075

In each washing the OD value of effluent was calculated and after the 3rd washing the least OD (0.032) value was recorded in the case of dyeing when *T. chebula* was used as mordant. The least leach out of the dye from the fabric in this dye-mordant combination indicates good color fastness and possible use of such combinations in natural dyeing process.

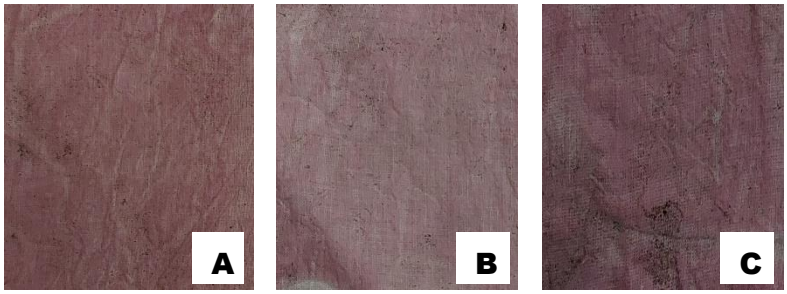

Fig 4.1: Cloth pieces dyed using Beetroot dye and various mordants; A – With mordant Terminalia, B – With mordant Carambola, C – With mordant embelica.

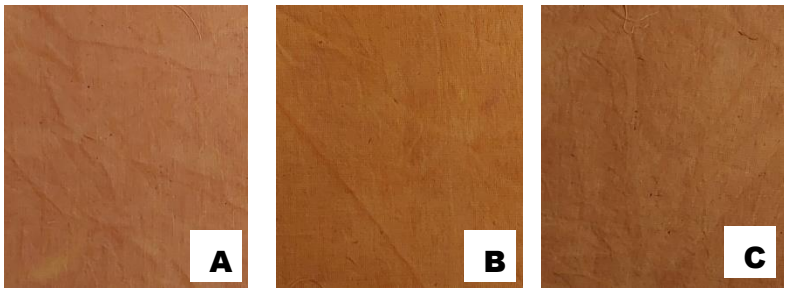

Fig 4.2: Cloth pieces dyed using Sappan wood dye and various mordants; A – With mordant Terminalia, B – With mordant Carambola, C – With mordant embelica.

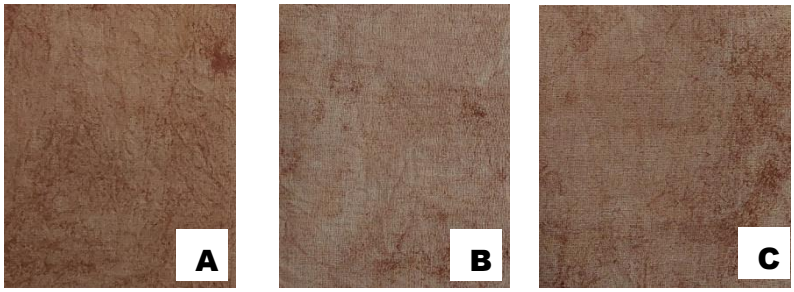

Fig 4.3: Cloth pieces dyed using Rubia dye and various mordants; A – With mordant Terminalia, B – With mordant Carambola, C – With mordant embelica.

5. DISCUSSIONS

5. DISCUSSIONS

Natural dyes are environmentally safe to wide applications including fabric dyeing. Therefore, utilization of such resources at a commercial level is to be promoted. However, need of developing dyeing techniques including the identification of suitable mordant dye combination to achieve various color shade is a prerequisite. In the present work such an attempt was made by using dye extract of *Beta vulgaris*, *Caesalpinia sappan*, and *Rubia cordifolia*. The selection suitable mordant is another important aspect in the fabric dyeing. In the present study we used three natural mordants *viz., Phyllanthus emblica*, *Terminalia chebula*, *Averrhoa carambola*. In the entire dyeing experiment, pre mordanting method was adopted. Cotton fabric, was destarched and used for the dyeing experiment.

Root tuber extract of *B. vulgaris* used for fabric and produced violet pink color to fabric by using *T. chebula* mordant. The stability of dye indicated that significant reduction in dye effluent in the third washing. Use of *B. vulgaris* in the fabric dyeing was reported earlier also (Yaqub et al., 2018). In the present study, the mature roots of *R. cordifolia* was used for dye extraction. The dyestuff produced deeper colours when it was dyed using *T. chebula* mordant. The color of the red/purple beetroot is due to a variety of betalain pigments. Some of the betalains in beet root dye are betanin, isobetanin, probetanin and neobetanin which are collectively caused betacyanin. Other pigments contained in beetroot are betraxanthins (Kujala et al., 2001; Azeredo et al., 2009). Red beet root contains both red and yellow pigments of the class betanines. These are quaternary ammonium amino acids. Red colour is at its most intense and variable in beetroot from deep reddish-purple to bright vermilion but some varieties also have orange yellow and white roots. These color differences are due to different levels of pigments (Betalains) that are characteristic of beetroot. (Kannan et al., 2011; Azeredo et al., 2009 Dadood et al., 2001). Beetroot red or betanin is used as a food coloring while beetroot juice is used for fabrics, leather and for hair colors (Lio, 1981; Kapoor et al., 2008;Qadariyah et al., 2018). Similarly, betalains, betacyanins and betaxanthins which are water soluble pigments are used as food colors, these also have antioxiadant properties (Kristen et al., 2006; Haddar et al., 2018). Furthermore, these natural dyes are rich in nutrients which are important for the human health (Nisa et al., 2006; Atamanova et al., 2007).

Caesalpinia sappan heartwood chips used for the dyeing cotton fabric produced vibrant reddish shades. *Phyllathus emblica* fruit extract when used as mordant, most bright color shade was achieved. When OD of effluent after 3rd washing analyzed spectrophotometrically, OD value recorded 0.032, indicating its high color fastness. Use of sappan wood extract as textile dye was reported earlier also (Ghorpade, 2000; Lee et al., 2008; Mitsunaga et al., 2009; Kannathasan and Kokila, 2021). However, fabric dyeing using sappan wood extract in combination with natural mordants like *P. emblica* fruit extract is the first report. The present result indicates its potential to use large scale fabric dyeing with safe, eco-friendly mordant like *Phyllanthus emblica* fruit extract.

The heartwood of *C. sappan* contains a red pigment called as "Brazilin" (De Oliveira et al., 2002). The plant has been found to contain medicinal properties such as antioxidant, antibacterial, anti-inflammatory and antiacne activities. Taif et al. (2019) used sappan extract to dyeing silk fabrics and reported that the intensity of color absorbed by silk fibre and its fastness properties against light. Boiling method used for the extraction of dye from the wood chips of sappan is in accordance Kannathasan and Kokila (2021). Mordanting and dyeing were conducted by pre-mordanting method found to be effective and is in conformity with the previous report (Mitsunaga et al., 2009). De Oliveira et al. (2002) reported that major pigment contained in crude extract of sappan wood is brazilin, which is classified as a neoflavanoid. This pigment, appear colorless or yellow color in nature, and is highly sensitive to air and light. Ohama and Trumpat (2014) reported that the cotton fabric dyed using sappan wood extracts without mordant had a shade of reddish brown, while those post-mordant with aluminium potassium sulphate, Ferrous sulphate and Copper sulphate produced a variety of wine red to dark purple color shades. In the present study, we observed colour strength was enhanced with the type of mordant used and was high in case of goose berry extract. Ohama and Tumpat (2014) reported the extraction method which contained 1:3 of sappan wood: water which comparable to the present study. In most of the treatments, the cotton fabrics showed fair wash fastness in our study.

Nirmal et al. (2015) reported that, Brazilin is the safe natural compound having potential to develop as a medicinal compound with application in food, beverage, cosmetics and

pharmaceutical industries to screen its clinical use in modern medicine. He also reported that the sappan wood contains various structural types of phenolic compounds including xanthone, coumarin, chalcones, flavones, homoisoflavonoids and brazilin, etc. This study support that when sappan dye is used for textile, this may additionally influence antibacterial activity through fabrics.

Rubia cordifolia aqueous root extract was used for the cotton fabric dyeing. The cotton dyed with *R. cordifolia* extract by pre mordanting method using *Terminalia chebula* fruit extract as mordant showed reddish brown color. The dyed cloth up on examination of its color fastness showed OD value of 0.032 after 3rdwashing effluent. *R. cordifolia* belongs to family Rubiaceae. The roots contain coloring component, anthraquinones. The chief colouring compounds isolated from rootsincludes alizarin (1, 3-dihydroxy-2-ethoxymethyl-9, 10-anthraquinone),purpurin (1,2,4-trihydroxy anthraquinone), xanthopurpurin (1,3- dihydroxyanthraquinone), munjistin [(1,3-dihydroxy-9,10-dioxo-9,10-dihydroanthracene-2-carboxylicacid (Mischenko et al., 1999), and pseudopurpurin(1,2,4-trihydroxy-9,10-dioxoanthracene-2-carboxylic acid) of which purpurin, xanthopurpurin and its methyl esters, and pseudopurpurin (purpurin 3-carboxylic acid) are only reported in tribe of *Rubia* (Swain, 1966).Naturally cotton fibers show reluctance to absorb natural dyes because of its cellulosic nature. In a preliminary trial, anthraquinone dye extract of Indian madder showed less affinity to cotton fibers. To enhance the affinity, different natural mordants were used, which also influenced the fastness property of dye on fabrics. As per the earlier reports (Samanta et al., 2010; Samanta et al., 2011) it is possible to get maroon red shade on jute fabric dyeing using Indian madder at alkaline medium. In a previous work (Bhuyan et al., 2004) anthraquinone and flavonoid moieties of native plants to dye natural silk and cotton fabrics were used. The use of alum, copper sulphate and ferrous sulphate as mordants for madder dye reported to produce various shades of creamish brown, chocolate brown, pink respectively (Agarwal and Gupta, 2003). In the present study, these mordants could produce various shades of red and purple color. It is also reported that madder dyed fabrics were brilliant shades of bright red to scarlet depending on mordants and dye concentrations (Yusuf et al., 2013).Good washing, perspiration fastness and fair light fastness of silk fabrics dyed with mixture of *R. cordifolia* and *Tagetus erecta* dye extracts was reported(Katti et al., 1996). Good or very good washing and moderate or fair light fastness was reported with mordants like copper sulphate, potassium dichromate or stannous chloride (Bhuyan et al., 2004); myrobalan and alum (Mondal

et al., 2004); ferrous sulphate and tannic acid (Teli et al., 2004); alum, copper sulphate, ferrous sulphate and myrobalan (Patel, 2011); tin (II) chloride (Yusuf et al., 2015) were used. The color fastness with respect to light exposure, washing and rubbing was quite satisfactory on woolen yarn when henna and madder dyed samples were analyzed (Yusuf etal., 2013). A comparative study on dyeing of cotton and silk fabric with madder by using alum and copper sulphate as mordants reported lesser wash fastness and somewhat similar rub fastness value (Jahan and Datta, 2015) as in the present investigation. The whole process of extraction and dyeing of madder that is used for the present work is ecologically safe and cost-effective. Since dye content in intact plants varies with season, age, soil type, processing methods etc…, special care is, however, required while selecting the root material to reproduce the same result as such. Brilliant colour shades obtained from Madder with different natural mordants will definitely attract the attention of the textile industry. In the present investigation madder root extract dyed by mordanting T. chebula extract through pre mordanting method was most promising for diverse reddish color shades. It was also shown that these mordants can improve the fastness property of the textile (Gulrajani, 2001).

In brief, various natural dye and natural mordant combinations tested by adopting pre mordanting method is suitable to achieve varying color shades with significant color fastness. However, further analysis of dyed fabric is needed to translate such combinations to industrial level dyeing purposes. Where dye stability, light stability, rub fastness etc. need to be accessed.

6. SUMMARY AND CONCLUSIONS

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Dyes are one of the natural products obtained from plant, animal and mineral sources. Majority of the natural dyes are obtained from plant kingdom. Per annum nearly 10, 00,000 tonnes of synthetic dyes are produced from various petroleum sources for various purposes. This vast amount of synthetic dyes cause environmental issues such as soil and water pollution consequent disturbance of the ecological balance and various health hazards to human beings and other organisms. The only alternative way to reduce the problems regarding synthetic dyes is the promoting the use of natural dyes. In the present study focus was mainly on the use of natural dyes from locally available plant materials like *Beta vulgaris*, *Caesalpinia sappan*, and *Rubia cordifolia*. The use of natural mordants from plant materials like *Phyllanthus emblica, Terminalia chebula,* and *Averrhoa carambola* were also studied.

The efficiency of mordants in cotton cloths was determined by using UV-Vis spectrophotometer. The absorption maxima of the dye solution derived from selected plant materials was recorded and OD of the dye solution before and after dyeing, color fastness and reflectance value were recorded on the basis of absorption maxima. Based on OD values the ability of different mordants to fix the natural dye to the fabrics was evaluated.

Present study revealed that pre - mordanting method of dyeing using of *C. sappan* wood extract with *P. embelica* mordant showed good dye-mordant combinations, and it gave least OD value. *T. chebula* was found to be a good mordant as it gave lowest OD value as compared to the other two mordants used.

B. vulgaris was found to be the most unstable dye as the cotton cloth didn't retain much of its color even in all three mordant – dye combinations used.

The present study was to explore the possibility of natural dyes as a main stream dyeing source. Many of the plant materials that are easily available can be used as source of dyeing, even many of the invasive species like *Clidemia hirta,Lantana camara* etc. can make a good source of natural dyes there by helping to control their spread. Even some of the plant waste materials like the peel of unripe banana (*Musa acuminata),* pineapple (*Ananas comosus*) are the major agro waste from our state that can be utilized for its high tannin containing a mordant. The

availability of high biomass and extractable phyto constituents from such candidates can be utilized as dyeing and mordant source in the field of textile industry. Natural dyeing is an eco-friendly and green technology ensures environmental safety, generate employment to rural population. Such innovations and ideas are a very important aspect for sustainable development and to reduce potential environmental and health hazard arising from using chemical or azo – based dyes.

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